STUDIES ON METHODS AND SYSTEMS FOR MEASURING RADON PROGENIES IN THE ATMOSPHERE

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Michio OKINO

Muroran Institute of Technology

September 1996

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By

Michio OKINO

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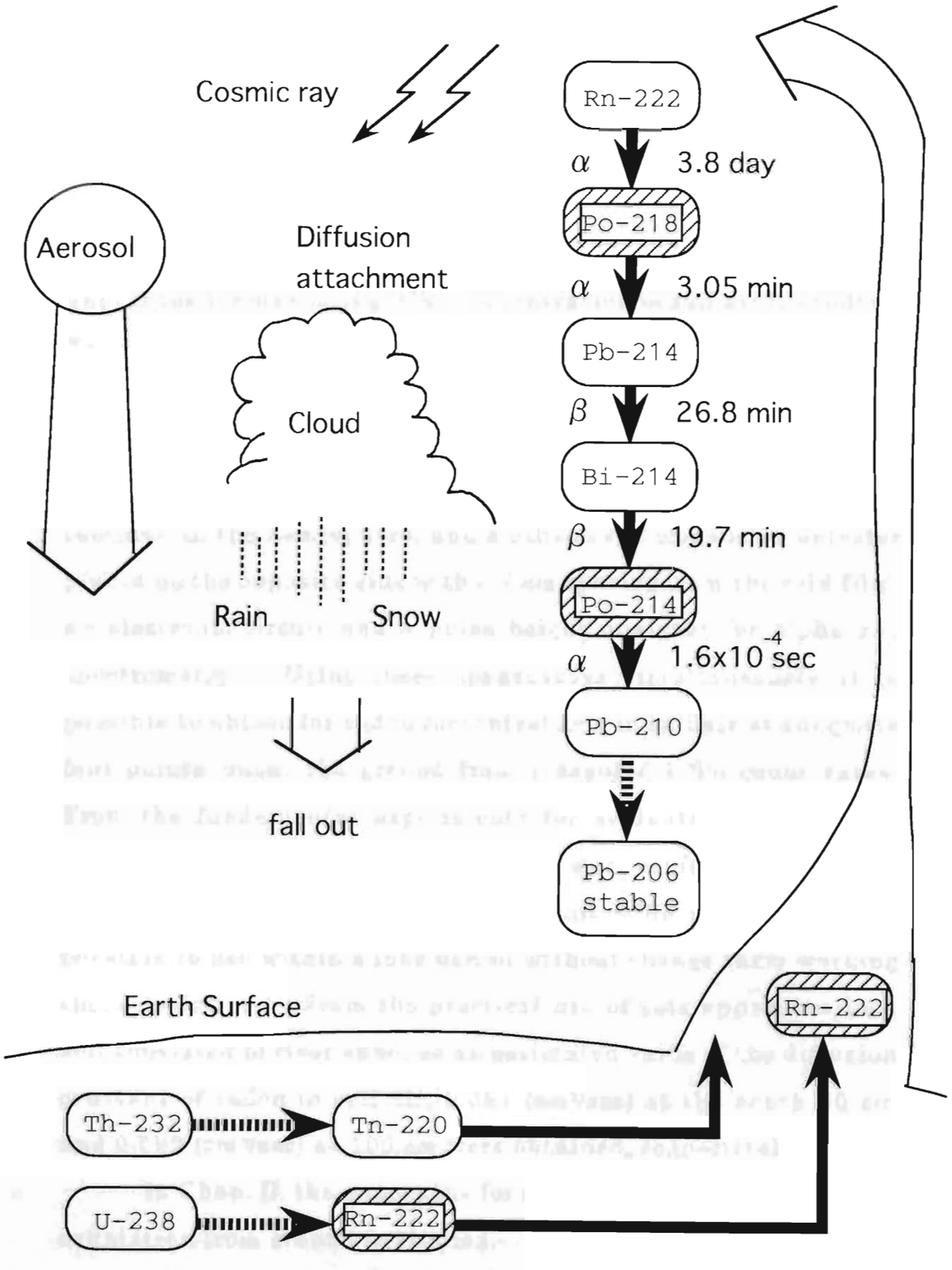
ABSTRACT



Schematic representation on the branches of present work.
- production of radon and its progeny, and their
circulation in the atmosphere -

ABSTRACT

This paper consists of four chapters. In Chap. I, the apparatus for measuring ^{222}Rn concentration in soil air is studied. To measure the concentration of ^{222}Rn in soil air, an apparatus has been developed and same four apparatuses were constructed. The apparatus is composed of a hemisphere of double screen meshes of 10 cm in diameter, gold film of 1.0 cm in diameter centered in the hemisphere, and a silicon semiconductor detector placed on the opposite side with 0.2 cm spacing from the gold film, an electronic circuit and a pulse height analyzer for alpha ray spectrometry. Using these apparatuses simultaneously, it is possible to obtain for radon concentrations in soil air at adequate four points under the ground from measured ^{218}Po count rates. From the fundamental experiments for evaluating the working characteristics of the apparatus, it was confirmed that the apparatus is useful to detect and count ^{218}Po in soil air, and possible to use within a long period without change their working characteristics. From the practical use of this apparatus in a soil consisted of river sand, as an estimated value of the diffusion constant of radon in soil air, 0.081 (cm^2/sec) at the depth 30 cm and 0.092 (cm^2/sec) at 100 cm were obtained, respectively.

In Chap. II, the apparatus for continuously measuring ^{222}Rn exhalation from ground is studied. To measure the rate of ^{222}Rn exhalation from the ground surface, an apparatus has been developed. The apparatus is composed of a radon collector, a 60-l cylindrical buffer tank, three kinds of filter and an ionization

chamber of flow-through type. To know the working characteristics and the accuracy of measured values, calibration measurements and comparison between values obtained with the present method and with other methods were made by applying two-filter method and by using activated charcoal method, respectively. From the basic experiments, the apparatus is capable of continuously recording measured data that provide an evaluation of ^{222}Rn exhalation rate with sufficient accuracy. From the practical use of the apparatus on the different ground conditions, following informations were obtained ; a) The apparatus operates stably even in stormy weather. b) From the measurements made on the ground inside of a house, the variation trends of exhalation rate were found to be in phase with the variations of temperature. c) From the measurements made on natural ground, the values on one day was considerably different from those at the same time on the other day. The variation of ground condition affected with the weather seems to be main cause to effect the variation of exhalation rates.

In Chap. III, the apparatus for measuring ^{222}Rn progeny concentration in atmosphere is studied. To measure the concentration of ^{222}Rn progeny in atmosphere, an apparatus has been developed that consists of a radon progeny collector, a semiconductor detector and a pulse height analyzer. A membrane filter (TOYO-ROSHI, Ltd. TM-300) was adopted for ^{222}Rn progeny collection. A silicon semiconductor detector (HORIBA, Ltd. 300SB 60L) was adopted for alpha ray detection of ^{222}Rn progenies collected on the filter. Alpha ray spectrometry

was adopted for the measurements of ^{218}Po and ^{214}Po. As a counting efficiency of this apparatus, a value 0.459 was obtained by calculation. A relative error was estimated to be within 20.4 percent.

From the observations made on the atmosphere over the ocean, it was confirmed that the apparatus is useful to measure extremely low level concentrations of ^{222}Rn progeny with sufficient accuracy.

From the observation made in the atmosphere over the Indian Ocean, followings are obtained ; a) On the mid Indian Ocean, radon concentration levels were found to be ranged from 6.6 $\times 10^{-4}$ Bq/m^3 to 7.6 $\times 10^{-2}$ Bq/m^3. b) Clear inverse correlation between radon concentration and electrical conductivity was found in the atmosphere over the mid Indian Ocean.

In Chap. IV, the apparatus for measuring the concentration of unattached ^{218}Po in atmosphere is studied. An apparatus has been developed that consists of three kinds of ^{218}Po collector provided with semiconductor detectors in which one of collectors is used for attached ^{218}Po measurement and others are for unattached ^{218}Po, and three pulse height analyzers. A 300 mesh wire screen was adopted for collection of unattached ^{218}Po. A membrane filter (TOYO-ROSHI, Ltd. TM-100) was adopted for collection of attached ^{218}Po. Silicon semiconductor detector (HORIBA, Ltd. 300SB 120L) were used for alpha ray detection. As collection efficiency of the wire mesh filter and appearing efficiency of ^{218}Po collected on the filter, values 0.72 and 0.686 were adopted in present work, respectively.

From some measurements carried out in a basement air, it was confirmed that the apparatus is capable of providing an evaluation of the unattached ^{218}Po concentration and the ratio of unattached ^{218}Po to attached ^{218}Po, simultaneously.

Chapter I

An apparatus for measuring ^{222}Rn concentration in soil air

I. An Apparatus for Measuring ^{222}Rn Concentration in Soil Air

1-1. Introduction

For measuring the concentration of radon in the soil air without disturbing natural condition of the soil, an apparatus has been developed on the basis of Mochizuki's method[1] and refering Jaki, et al[2]., and same four apparatuses were constructed.

The apparatus is composed of a hemisphere of double screen meshes of 10 cm in diameter, a gold film of 1.0 cm in diameter centered in the hemisphere, and a silicon surfaced barrier type semiconductor detector (S.S.D.) placed on the opposite side of gold film with 0.2 cm spacing from the gold film, and an electronic circuit for alpha ray spectrometry. Specifications of S.S.D. used are : 0.5 cm^2 in surface area, 60 μm in effective thickness and 48.9 keV in resolution.

^{218}Po flowing into effective volume of the hemisphere is collected on the gold film by the aid of electric field between the inner mesh and the gold film. Alpha ray emitted from the radioactive substances that have been collected on the gold film is detected and counted by the aid of S.S.D. placed at opposite side of the gold film.

By this apparatus, radon concentration in the soil air at adequate points inner the depth of one meter under the ground is obtained from the concentration of ^{218}Po which was measured in the same air. Further, it is possible to obtain the exhalation rate of radon from the variation state and decreasing rate of radon concentration which were measured at each points under the

ground.

1-2. Method of Measurement and Structure of Apparatus

In the soil air, it is thought that radioactive equilibrium state between ^{222}Rn and ^{218}Po is normally established. Therefore, ^{222}Rn concentration at adequate depth under the ground is obtained from the concentration of ^{218}Po which is measured directly in same soil air. Since ^{218}Po is positively charged when it produces, it can be collected by the aid of electric field.
Figure 1-1 shows a principle of the measurement. Figure 1-2 shows the structure of device and its dimensions.

^{218}Po which floats in effective volume of the hemisphere is collected on the gold film by the aid of electric field between the inner mesh and the film. Alpha ray emitted from the radioactive substances collected on the film is detected by S.S.D.

This device provided with S.S.D. to measure ^{218}Po concentration is composed of a hemisphere of double screen meshes, 10 cm in diameter, a gold film centered in the hemisphere, 1.0 cm in diameter and a S.S.D. placed behind gold film spacing of 0.2 cm. Heater, which dry up the insulator surfaces, also locates around the film. Characteristics of S.S.D. used are as follows: surface area is 0.5 cm^2, useful thickness is 60 μm and resolution is 48.9 keV. The voltage used as bias is 50 V. The quality of the material of device is consists of brass and acrylic resin plate. Electronic circuit used is shown in Fig. 1-3.

Negative voltage 140V which cause the electric field between the inner mesh and the film is supplied to the film by dry battery.

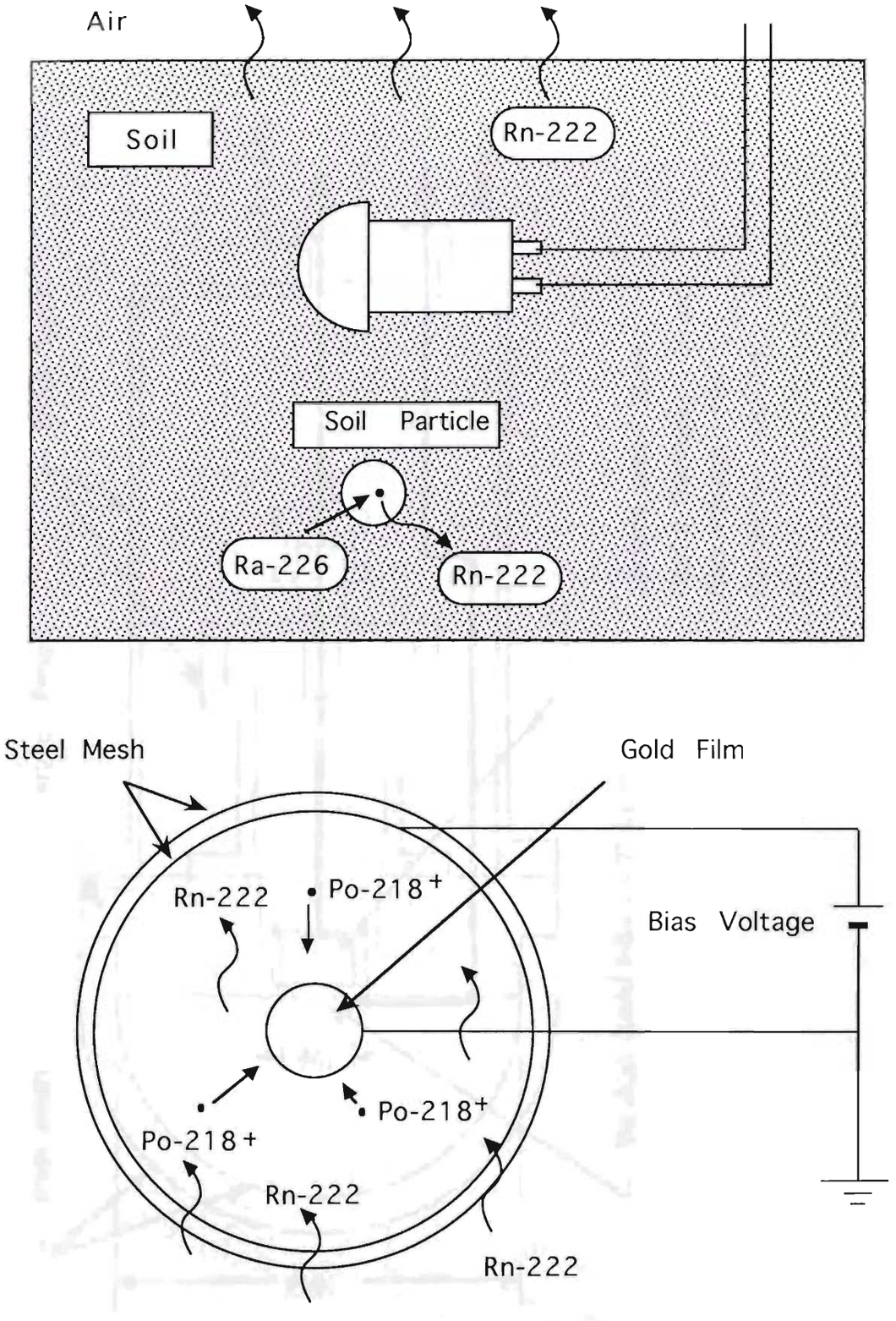
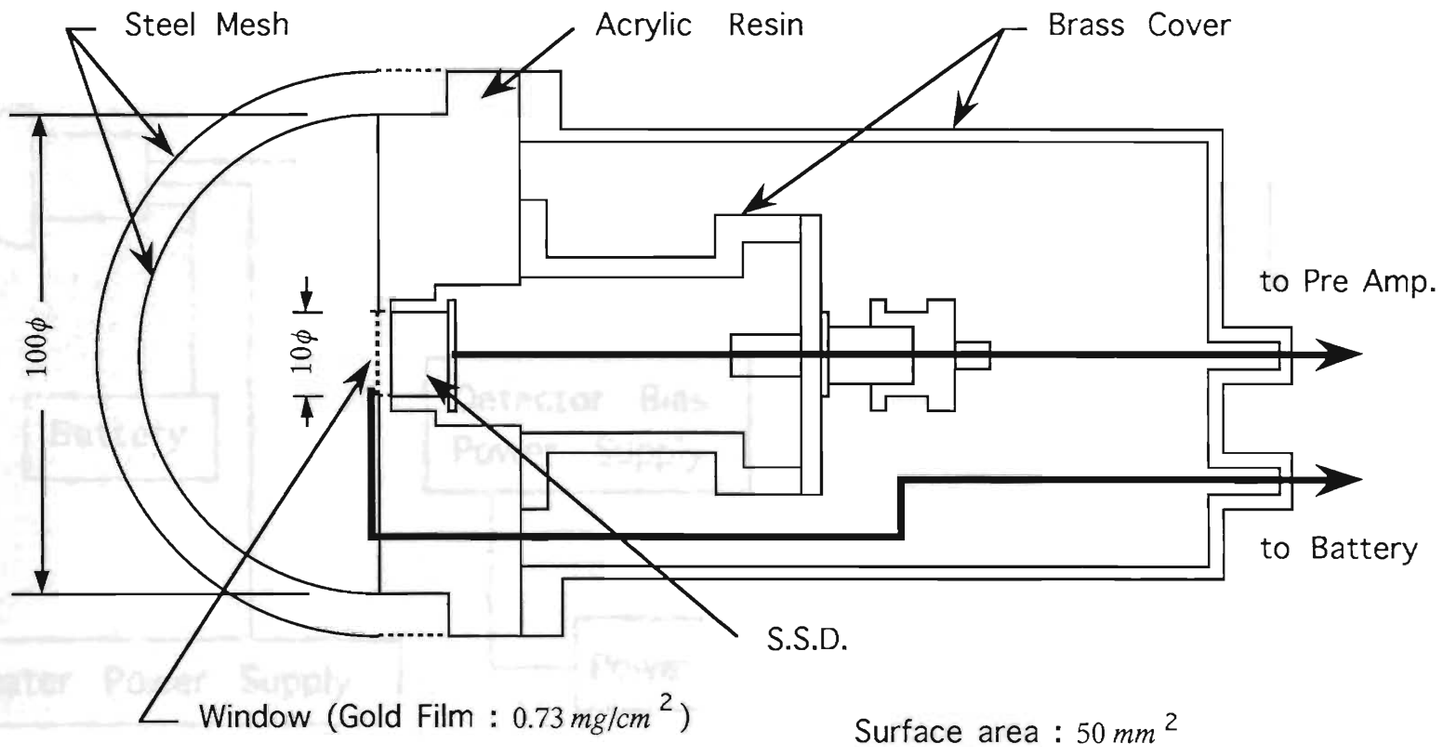


Fig. 1-1 Schematic representation of the measuring principle.



Surface area : 50 mm^2
Useful thickness : 60 μm
Bias voltage : 50 V
Resolution : 48.9 KeV

Fig. 1-2 Structure of the collecting device and its dimensions.

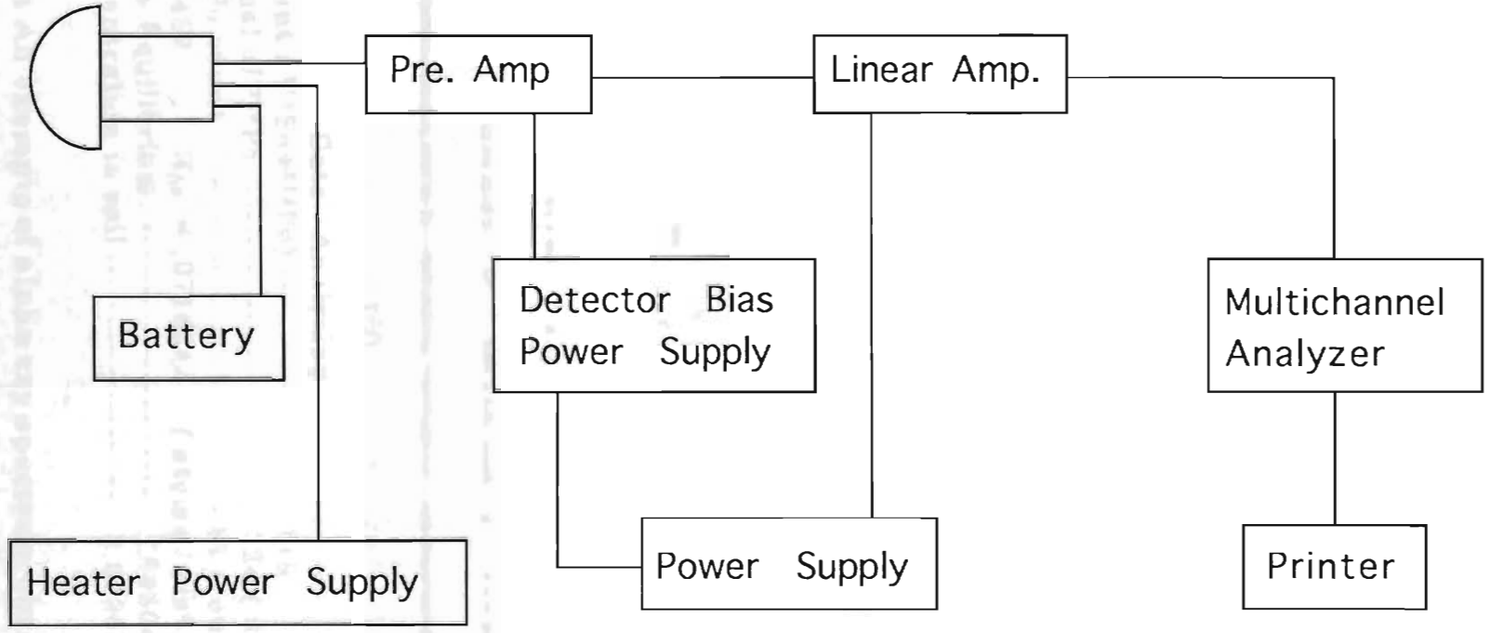
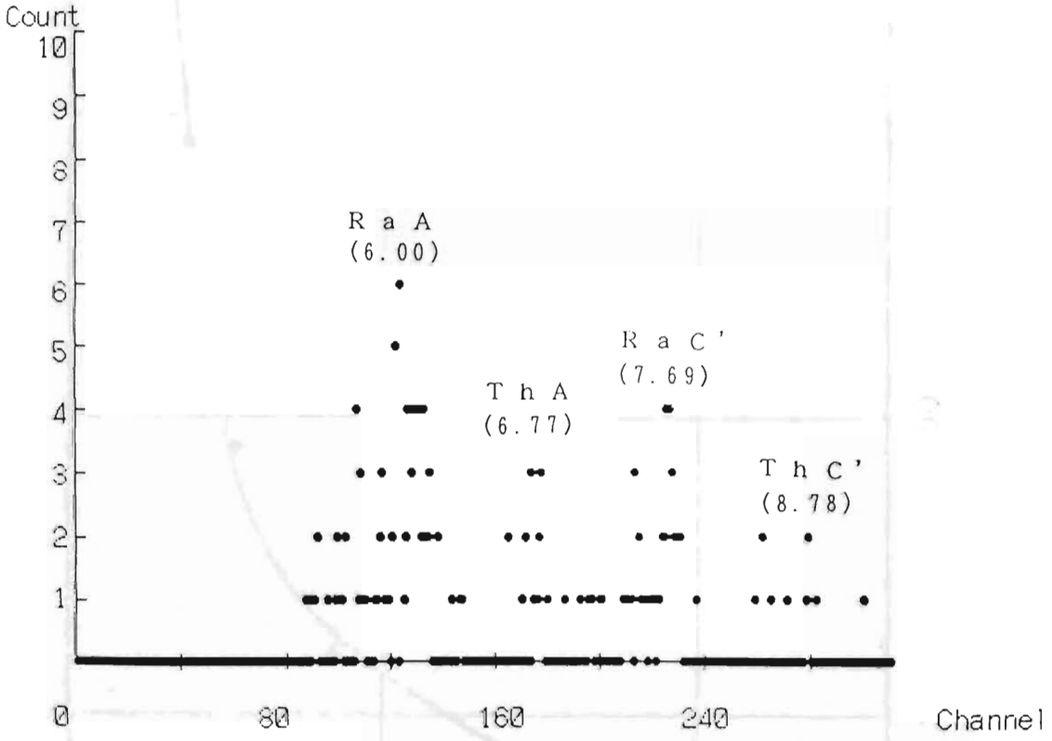


Fig. 1-3 Electronic circuit used to counting system for alpha ray spectrometry

---------- Device Setting ----------

14. Oct. 1994 in shed for obserbation

Device number ---------- No. 1
Sample ---------- River sand
Sampling time ---------- 8000 (sec)
Bias Voltage ---------- 45 (V)
Coarse Gain ---------- 8
Fine Gain ---------- 3-50
Gain ---------- 0-90
Shaping ---------- 0.8 (micro sec)
U.L. ---------- 10-10
L.L. ---------- 0-80



---------- Data Analyzing ----------

Sum of Count ($^{218}Po+^{214}Po$) ---------- 118
Peak channel of ^{218}Po ---------- 124 (6 count)
Sum of ^{218}Po count ---------- 81 (count)
N_{A1} = .186452 : N_{A2} = .0716542 (atoms / cm^3)
Radioactive Equilibrium ---------- .384304
^{222}Rn Concentration in soil ---------- 7.0736E-04 (Bq / cm^3)

Fig. 1-4 An example of alpha ray spectrum obtained from in air within a soil composed of river sand at the depth 100 cm

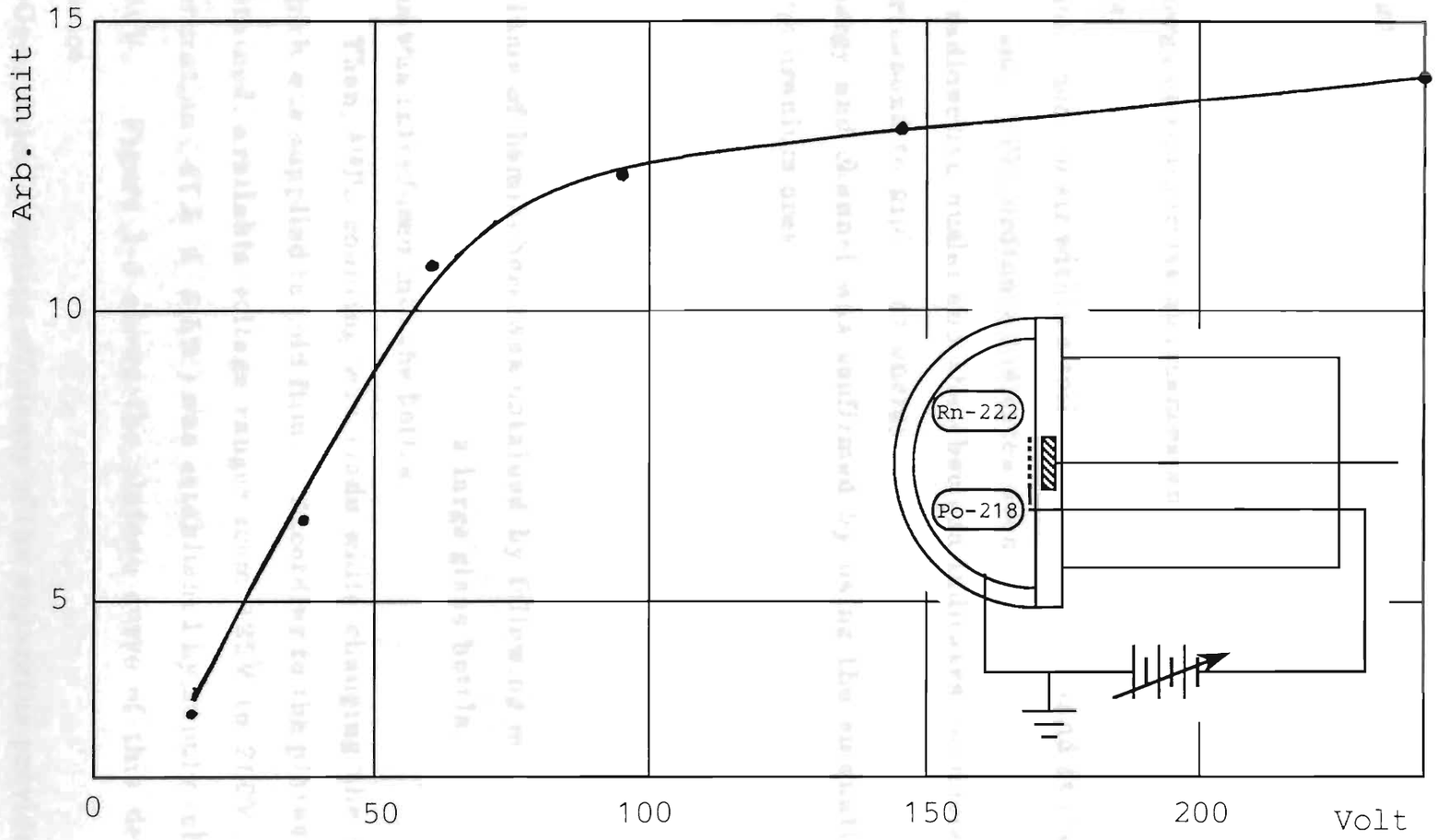


Fig. 1-5 Plateau curve of the collecting device

Signals from S.S.D. are fed to multichannel analyzer through pre amplifier, linear amplifier and biased amplifier. Digital data for each channel are printed out by a digital recorder. The relation among spectrum displayed on the visual display tube, energy of radioactive substances and channels are shown in Fig. 1-4. This alpha ray spectrum was obtained from radioactive substances in air within a soil composed of river sand at the depth 100 cm. The ordinate indicates events of alpha disintegration of radioactive nuclei and the abscissa indicates channels which correspond to alpha ray energy. The relation between each of energy and channel was confirmed by using the emanation gas from uranium ores.

1-2-1. Working Characteristics

a) The plateau curve

The plateau curves for collection of ^{218}Po in the effective volume of hemisphere was obtained by following manners ; The detecting device was placed in a large glass bottle. And radon gas was introduced into the bottle.

Then, ^{218}Po counting was made while changing the voltage which was supplied to gold film. According to the plateau curve obtained, available voltage ranges from 125V to 250V. The saturation (87.5 $\pm$ 5.5%) was established by supply voltage of 250V. Figure 1-5 shows the plateau curve of this detecting device.

b)Geometrical counting efficiency of the apparatus provided with S.S.D. First, suppose that following 1), 2), 3) and 4) are

presented ;

1) (a) shown in Fig. 1-A represents the window of S.S.D. and (b) filter, r and d represent their radius, respectively, and h represents the distance between them.

2)Co-ordinates, x and g, are given as shown in Fig. 1-B.

3)Radiation point source is placed at, C, keeping away the distance, x, from the center, O, of the filter.

4)Shaded portion on the unit sphere, 1 in radius, S_1 , shown in Fig. 1-B represents a spherical surface area which is made by projection of upper disk ((a) shown in Fig. 1-B (S.S.D. window)) to the lower one ((b) filter) as taking the focus at, C, on (b) shown in Fig. 1-B.

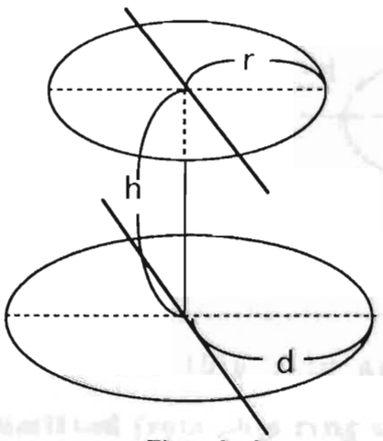


Fig. 1-A.

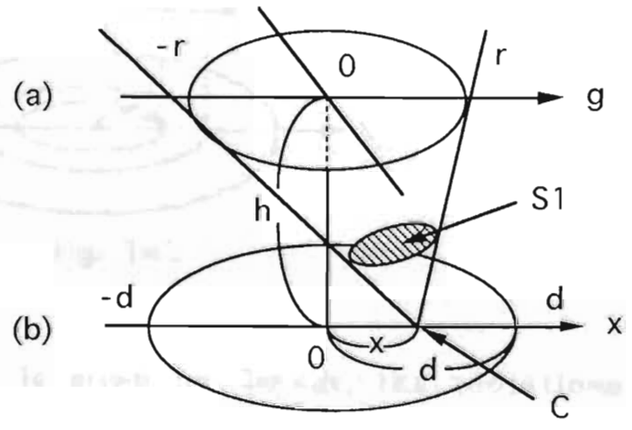


Fig. 1-B.

And assume that radiation absorption caused by air between S.S.D. window and the filter can be negligible.

Since a spherical surface area of the unit sphere, 1 in radius, is given by 4π, the counting efficiency, $G(x)$, will be given by

$$G(x) = \frac{S_1}{4\pi}$$

Taking into account, g, as a parameter, S_1 is expressed by

$$S_1 = 2\int_{-r}^{r} \frac{h}{h^2 + (x-g)^2} \sqrt{\frac{r^2 - g^2}{r^2 - g^2 + h^2 + (x-g)^2}}\, dg \qquad (1\text{-}1)$$

Second, assume that the radioactive substances are uniformly distributed on the filter. Let's denotes, I, the total radiation emitted from the radioactive substances on the filter.

Suppose that a ring, dx in width, is presented on the filter as shown in Fig. 1-C.

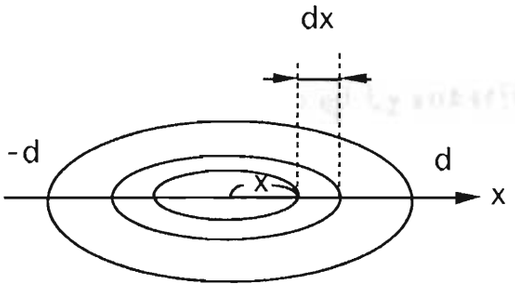


Fig. 1-C.

Since, this ring area is given by $2\pi x \times dx$, the radiations emitted from this ring area are given by

$$I \times \frac{2\pi x}{\pi d^2} dx$$

In this occasion, the radiations entering through the S.S.D. window will be given by

$$I \times \frac{2\pi x \cdot dx}{\pi d^2} G(x) \qquad (1\text{-}2)$$

Thus, the counting efficiency $[G]$ for radioactive substances distributed uniformly on the filter will be given by integrating (1-2) from 0 to d for x, and dividing it by I as follows ;

$$[G] = \int_0^d \frac{2\pi x \cdot dx}{\pi d^2} G(x)$$

$$[G] = \frac{1}{\pi d^2} \int_0^d \int_{-r}^{r} \frac{hx}{D(x)} \sqrt{\frac{E}{E + D(x)}} \, dg \cdot dx \qquad (1\text{-}3)$$

where,

$$D(x) = h^2 + (x - g)^2 , \qquad E = r^2 - g^2$$

A value of $[G]$, 0.0625 was obtained by substituting, $d = 5$, $r = 4$, $h = 3$ into the equation (1-3).

$[G]$ for various values of d, r and h was calculated.

c) Examination in the soil air modeled upon the soil circumstance

Since the device is used in the soil under the ground, it is necessary that the working characteristics are not changed in the soil circumstance where the humidity is considered to be high. Therefore, the working characteristics of device was examined under the condition that the device was placed in the bottle mentioned above and was covered by the soil. Under this condition, counting state of ^{218}Po was checked on sometimes by introducing the radon gas into the soil in the bottle.

From fundamental experiments carried out over long duration,

^{218}Po in the soil air, and further, the apparatus is possible to use within a long period of time without change of its working characteristics in the soil circumstance.

1-3. Deriving the ^{222}Rn Concentration from the ^{218}Po Counting Rate

Following consideration is adopted when the detector device was placed in a adequate depth under the ground.

Used notations are as follows ;

n_{Rn} : Concentration of ^{222}Rn atoms in air in effective volume surrounded by the inner mesh [atoms/cm^3]

n_A, n_B, n_C : Concentration of ^{218}Po, ^{214}Pb and ^{214}Bi atoms in air in effective volume surrounded by the inner mesh, respectively [atoms/cm^3]

N_A, N_B, N_C : Number of ^{218}Po, ^{214}Pb and ^{214}Bi atoms on the film, respectively [atoms]

$I_{\alpha 1}$: Alpha disintegration rate of ^{218}Po on the film at time t [dps]

$I_{\alpha 2}$: Alpha disintegration rate of ^{218}Po and ^{214}Po on the film at time t [dps]

$\lambda_{Rn}, \lambda_A, \lambda_B, \lambda_C$: Decay constant of ^{222}Rn, ^{218}Po, ^{214}Pb and ^{214}Bi, respectively [sec^{-1}]

V: Effective volume surrounded by the inner mesh [cm^3]

ε : Counting efficiency of the detecting device for radiation of radioactive substances on the film

C : Collection efficiency of film for radioactive aerosols

C : Collection efficiency of film for radioactive aerosols

t : Collection time [sec]

The time variation of n_A in the effective volume is expressed by the following differential equation ;

$$\frac{dn_A}{dt} = \lambda_{Rn} n_{Rn} - \lambda_A n_A \tag{1-4}$$

putting $n_A = n_{A0}$ at $t = 0$,

$$n_A = \frac{\lambda_{Rn} n_{Rn}}{\lambda_A} + \left(n_{A0} - \frac{\lambda_{Rn} n_{Rn}}{\lambda_A} \right) e^{-\lambda_A t} \tag{1-5}$$

For $t >$ about 2000, the equation (1-5) reduces to

$$n_A = \frac{\lambda_{Rn} n_{Rn}}{\lambda_A} \tag{1-6}$$

This equation express radioactive equilibrium between ^{222}Rn and ^{218}Po atoms. During the collection period, the time variation of N_A on the film may be expressed by the following equation ;

$$\frac{dN_A}{dt} = CV\lambda_A N_A - \lambda_A N_A \tag{1-7}$$

Solving this equation by means of the Laplace transformation,

$$sN_A(s) - N_A(0) = \frac{CV\lambda_A N_A}{s} - \lambda_A N_A(s)$$

$$N_A(s) = \frac{CV\lambda_A N_A}{s(s+\lambda_A)} + \frac{N_A(0)}{s+\lambda_A}$$

$$= CVN_A\left(\frac{1}{s} - \frac{1}{s+\lambda_A}\right) + \frac{N_A(0)}{s+\lambda_A}$$

where s is parameter.

Initial condition is $N_A = 0$ at $t = 0$. Consequently, the solution of the equation is as follows :

$$N_A = CVn_A\left(1 - e^{-\lambda_A t}\right) \qquad (1\text{-}8)$$

$$= CVn_A f_1(t) \qquad (1\text{-}8')$$

$I_{\alpha 1}$ may be expressed by the following equation :

$$I_{\alpha 1} = \lambda_A N_A \qquad (1\text{-}9)$$

Inserting equation (1-8) into (1-9), $I_{\alpha 1}$ is

$$I_{\alpha 1} = CV\lambda_A n_A\left(1 - e^{-\lambda_A t}\right) \qquad (1\text{-}10)$$

The relation between $I_{\alpha 1}$ and $I_{\alpha 1}'$ is

$$I_{\alpha 1}' = \varepsilon I_{\alpha 1} \qquad (1\text{-}11)$$

where $I_{\alpha 1}'$ is net counting of ^{218}Po.

Inserting equation (1-10) into (1-11), n_A is

$$n_A = \frac{I_{\alpha 1}'}{\varepsilon C V \lambda_A \left(1 - e^{-\lambda_A t}\right)} \qquad (1\text{-}12)$$

Expressing n_A in Ci unit, ^{218}Po concentration in the soil C_{RaA} is given by

$$C_{RaA} = \frac{\lambda_A n_A}{3.7 \times 10^{10}} = \frac{I_{\alpha 1}'}{\varepsilon C V \left(1 - e^{-\lambda_A t}\right) \cdot 3.7 \times 10^{10}} \qquad (1\text{-}13)$$

Assuming radioactive equilibrium being held between ^{222}Rn and ^{218}Po, n_{Rn} is

$$n_{Rn} = \frac{\lambda_A}{\lambda_{Rn}} \cdot n_A \qquad (1\text{-}14)$$

Expressing n_{Rn} in Ci unit, ^{222}Rn concentration C_{Rn} is given by

$$C_{Rn} = \frac{\lambda_{Rn} n_{Rn}}{3.7 \times 10^{10}} \qquad (1\text{-}15)$$

Inserting equation (1-14) into (1-15), hence

$$C_{Rn} = \frac{\lambda_A n_A}{3.7 \times 10^{10}} = C_{RaA} \qquad (1\text{-}16)$$

Therefore, assuming radioactive equilibrium between ^{222}Rn and ^{218}Po in effective volume, ^{222}Rn concentration is equal to ^{218}Po concentration. Thus, ^{222}Rn concentration in the soil air is given by the following equation

$$C_{Rn} = \frac{I_{\alpha 1}'}{\varepsilon CV\left(1-e^{-\lambda_A t}\right)\cdot 3.7\times 10^{10}} \qquad (1\text{-}17)$$

Expressing C_{Rn} in Bq unit, Rn concentration C_{Rn} is given by

$$C_{Rn} = \frac{I_{\alpha 1}'}{\varepsilon CV\left(1-e^{-\lambda_A t}\right)} \qquad (1\text{-}17')$$

Evaluation of equilibrium state and estimation of relative error

If radioactive equilibrium state is being held like as $\lambda_{Rn} n_{Rn} = \lambda_A n_A$ and $\lambda_{Rn} n_{Rn} = \lambda_A n_A'$, two concentrations for n_{Rn} values, $(\lambda_A/\lambda_{Rn})n_A$, $(\lambda_A/\lambda_{Rn})n_A'$ were obtained. So that the ratio n_A/n_A' should be equal to 1. Thus, the observed ratio is useful to determine the fraction of secular equilibrium.

A relative error, $\Delta R/R$, calculated from $R = n_A'/n_A$ is given by the equation,

$$\left|\frac{\Delta R}{R}\right| = \left|\frac{En_A}{n_A}\right| + \left|\frac{En_A'}{n_A'}\right|$$

where En_A and En_A' are the errors of n_A and n_A' , respectively.

As described above,

$$n_A = \frac{I_{\alpha 1}'}{\varepsilon CV\lambda_A f_1(t)} \quad , \qquad n_A' = \frac{I_{\alpha 2}'}{\varepsilon CV\lambda_A f_2(t)}$$

where ε , C , $I_{\alpha 1}{}'$ are the measuring values, and V , λ_A , $f_1(t)$, $f_2(t)$ are the constant values. Therefore, the probable errors, En_A ,of n_A and $En_A{}'$, of $n_A{}'$ are given as follows ;

$$\frac{\partial n_A}{\partial \varepsilon} = -\frac{I_{\alpha 1}{}'}{\varepsilon^2 C V \lambda_A f_1(t)} \tag{1-18}$$

$$\frac{\partial n_A}{\partial V} = -\frac{I_{\alpha 1}{}'}{\varepsilon C V^2 \lambda_A f_1(t)} \tag{1-19}$$

$$\frac{\partial n_A}{\partial I_{\alpha 1}{}'} = -\frac{1}{\varepsilon C V \lambda_A f_1(t)} \tag{1-20}$$

therefore, En_A is obtained as,

$$En_A{}^2 = \left(\frac{\partial n_A}{\partial \varepsilon}\right)^2 \varepsilon_\varepsilon{}^2 + \left(\frac{\partial n_A}{\partial C}\right)^2 \varepsilon_C{}^2 + \left(\frac{\partial n_A}{\partial I_{\alpha 1}{}'}\right) \varepsilon_{I_{\alpha 1}{}'}{}^2$$

or

$$\left(\frac{En_A}{n_A}\right)^2 = \left(\frac{\varepsilon_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\varepsilon_C}{C}\right)^2 + \left(\frac{\varepsilon_{I_{\alpha 1}{}'}}{I_{\alpha 1}{}'}\right)^2 \tag{1-21}$$

and for $n_A{}'$

$$\left(\frac{En_A{}'}{n_A{}'}\right)^2 = \left(\frac{\varepsilon_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\varepsilon_C}{C}\right)^2 + \left(\frac{\varepsilon_{I_{\alpha 2}{}'}}{I_{\alpha 2}{}'}\right)^2 \tag{1-22}$$

where ε_ε , ε_C , $\varepsilon_{I_{\alpha 1}{}'}$ and $\varepsilon_{I_{\alpha 2}{}'}$ are the probable errors of ε , C , $I_{\alpha 1}{}'$

and $I_{\alpha 2}'$, respectively.

The maximum deviation from the mean of the experimentally determined values for these errors of the instruments amounted to ± 3 percent, ± 6.5 percent, ± 2.6 percent and ± 12.2 percent for ε, C, $I_{\alpha 1}'$ and $I_{\alpha 2}'$, respectively.

Now, substituting these values to the equation (1-21) and (1-22), values of En_A/n_A and En_A'/n_A' are obtained as

$$\frac{En_A}{n_A} = \pm 7.0 \qquad \text{and} \qquad \frac{En_A'}{n_A'} = \pm 13.8 .$$

Therefore, the results obtained for $\Delta R/R$ were certainly correct to within 20.8 percent.

1-4. Practical Application of the Apparatus

Trial estimation on the diffusion constant of radon in soil air.

Let C_S be the concentration of ^{222}Rn in the soil air (atoms/cm^3), d its diffusion constant (cm^2/sec), and a its rate of production with in soil (atoms/cm^3 · sec), which can be assumed to be independent of depth. Equilibrium conditions within the soil for the diffusion transport of ^{222}Rn toward the earth's surface can then be expressed by the following equation ; [3]

$$\frac{\partial C_S}{\partial t} = 0 = d\frac{\partial^2 C_S}{\partial z^2} + a - \lambda C_S \qquad (1\text{-}23)$$

where z is the depth, t the time, and λ the ^{222}Rn decay rate. The solution of (1-23) is

$$C_S = \frac{a}{\lambda}\left[1 - \exp\left(-\sqrt{\frac{\lambda}{d}} \cdot z\right)\right] \qquad (1\text{-}24)$$

where $C_{S0} = \frac{a}{\lambda}$ is the concentration of ^{222}Rn in undisturbed soil air in deeper layers. This gives an exhalation rate

$$E = \left(d\frac{\partial C_S}{\partial z}\right)_{z=0} = a\sqrt{\frac{d}{\lambda}} \qquad (1\text{-}25)$$

Accordingly, if ^{222}Rn concentration C_S at the depth z is known, values for d can be calculated from simultaneous equations introduced from the equation (1-24) where C_S and z are variables, a and λ are constants.

For obtaining the ^{222}Rn concentration in soil air with depth, the measurements were made using four apparatuses buried within a river sand at the depth 10 cm, 30 cm, 60 cm and 100 cm, respectively. Detail explanation on the river sand used and experimental condition were described in Chapter 3 Section 4. The results obtained are presented in Table 1-1.

For determining the rate of radon production within river sand, the concentration measurements were made using a device shown in Fig. 1-6. The value 1.74 $\times 10^{-9}$ (Bq/cm^3 · sec) as the rate of production a was obtained. Inserting values a 1.74 $\times$ 10^{-9} (Bq/cm^3 · sec) and λ 2.1 $\times 10^{-6}$ (sec^{-1}) as constants into the equation (1-24), diffusion constant d (cm^2 · sec) was calculated with correspond to the depth z (cm). The results obtained are presented in Table 1-2.

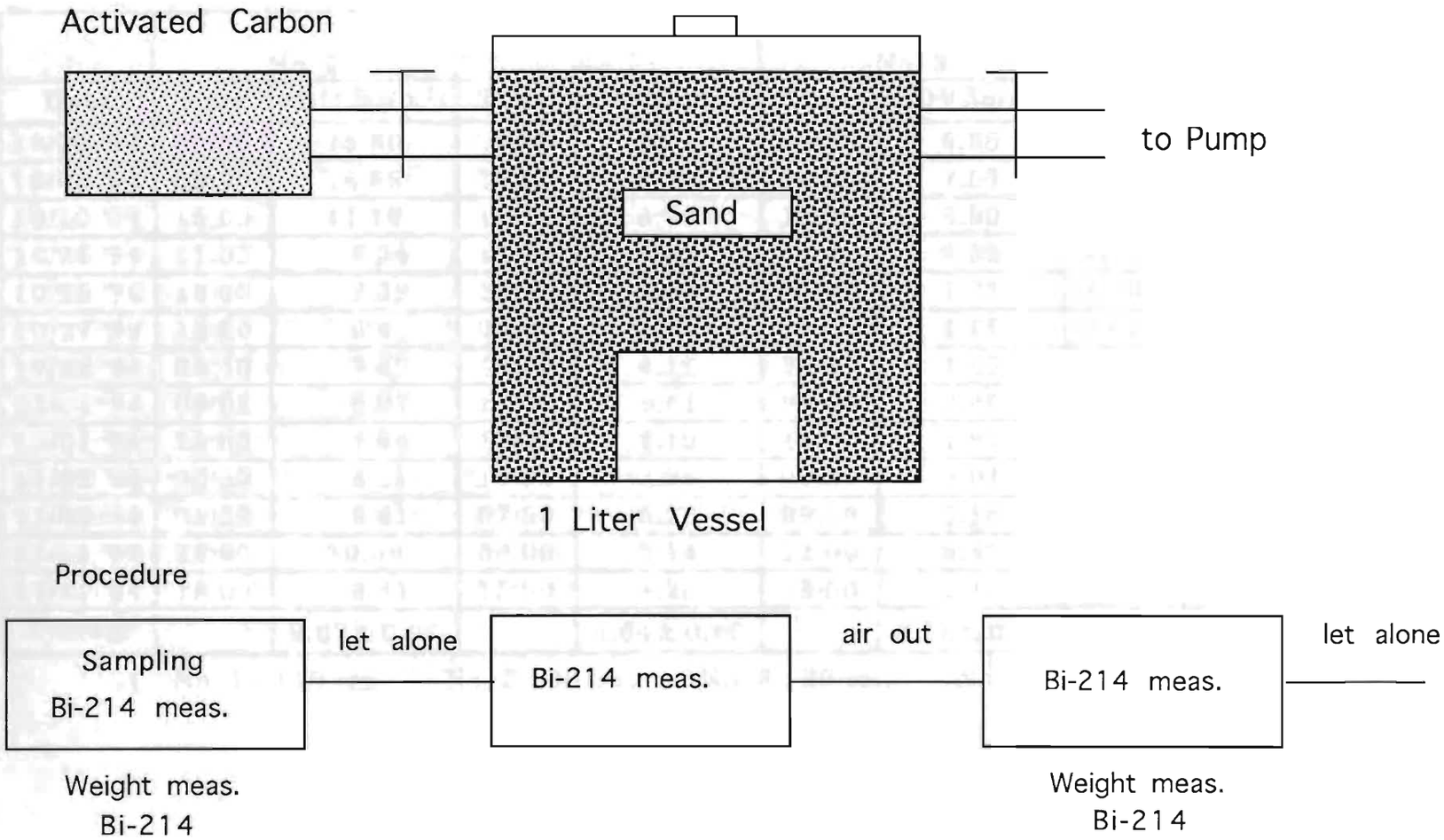


Fig. 1-6 Schematic representation for measuring escape rate of ^{222}Rn from sand particle into soil air.

	No. 1		No. 2		No. 3		No. 4	
Date	Time	x10²(Bq/m³)	Time	x10²(Bq/m³)	Time	x10²(Bq/m³)	Time	x10²(Bq/m³)
10/05 '94	09:30	15.80	12:00	8.91	09:30	5.30	12:00	3.29
10/06 '94	09:00	16.65	15:00	7.25	09:00	7.07	15:00	5.77
10/10 '94	15:00	11.10	12:10	5.76	15:00	5.30	12:10	2.49
10/26 '94	11:00	7.54	13:00	4.80	11:00	2.39	13:00	0.60
10/26 '94	16:00	6.69	18:30	2.97	16:00	1.25	18:30	1.49
10/27 '94	13:30	6.41	09:00	4.19	13:30	1.25	09:00	0.90
10/28 '94	09:40	7.83	14:00	4.19	09:40	1.97	14:00	0.90
11/01 '94	09:05	6.97	11:30	3.41	09:05	2.49	11:30	1.39
11/01 '94	14:00	7.54	17:00	4.19	14:00	1.97	17:00	1.69
11/08 '94	09:30	8.11	14:00	3.93	09:30	3.01	14:00	2.89
11/09 '94	09:10	6.41	07:00	3.23	09:10	2.18	07:00	2.09
11/11 '94	12:00	10.39	06:00	3.14	12:00	3.43	06:00	1.49
11/11 '94	18:00	6.41	15:00	4.28	18:00	2.18	15:00	1.39
Average		9.07±0.93		4.64±0.46		3.06±0.47		2.03±0.37

(No. 1 : 100 cm, No. 2 : 60 cm, No. 3 : 30 cm, No. 4 : 10 cm)

Tab. 1-1 Result of measurement of ^{222}Rn concentration in soil air at the depth 10 cm, 30 cm, 60 cm and 100 cm.

Date	No. 1 (cm^2/sec)	No. 2 (cm^2/sec)	No. 3 (cm^2/sec)	No. 4 (cm^2/sec)
10/05 '94	0.033	0.028	0.016	0.004
10/06 '94	0.031	0.039	0.010	0.002
10/10 '94	0.055	0.056	0.016	0.007
10/26 '94	0.101	0.077	0.067	0.105
10/26 '94	0.122	0.180	0.227	0.018
10/27 '94	0.131	0.097	0.227	0.048
10/28 '94	0.095	0.097	0.095	0.048
11/01 '94	0.114	0.140	0.062	0.020
11/01 '94	0.101	0.097	0.095	0.014
11/08 '94	0.090	0.109	0.044	0.005
11/09 '94	0.131	0.154	0.079	0.009
11/11 '94	0.061	0.162	0.035	0.018
11/11 '94	0.131	0.094	0.079	0.020
Average	0.092	0.102	0.081	0.024

(No. 1 : 100 cm, No. 2 : 60 cm, No. 3 : 30 cm, No. 4 : 10 cm)

Tab. 1-2 Diffusion constants in air of soil constructed from river sand.

As seen in Tab. 1-2, the average values seem considerably high compared with the corresponding value 0.05 (cm^2/sec) presented by Israël[4][5] except the value obtained at the depth 10 cm. However, it is considered that the method used in the present work is useful for evaluation of the diffusion constant of radon in soil air which is composed of various kinds of soil elements.

1-5. Summary and concluding remarks

For measuring the concentration of ^{222}Rn in the soil air, an apparatus has been developed. By this apparatus, ^{222}Rn concentration in the soil air at adequate points under the ground is obtained from ^{218}Po concentration measured without disturbing natural condition of the soil. The apparatus consists of a hemisphere of double screen meshes of 10 cm in diameter, a gold film of 1.0 cm in diameter centered in the hemisphere, a silicon surface barrier type semiconductor detector placed on the opposite side of the hemisphere, and an electronic circuit for alpha ray spectrometry.

To know the working characteristics of the device, some experiments and calculations were made.

The results obtained are follows ;

a) Negative voltage 140V for collecting ^{218}Po in the effective volume of hemisphere was determined from the plateau curve experiment.

b) A value 0.0625 as a geometrical counting efficiency was obtained from calculation.

c) Relative error was estimated as 20.8 percent when evaluation of equilibrium state is made.

d) The apparatus is useful to detect and count ^{218}Po in soil air and is possible to use within a long period without change of its working characteristics.

e) From the practical use of this apparatus in a soil consists of river sand, 0.081 (cm^2/sec) at the depth 30 cm and 0.092 (cm^2/sec) at 100 cm were obtained, respectively.

Chapter II

An apparatus for continuously measuring ^{222}Rn exhalation from ground

II. An Apparatus for Continuously measuring ^{222}Rn Exhalation from Ground

2-1. Introduction

For the purpose of continuously measuring the rate of ^{222}Rn exhalation from the ground surface, an apparatus has been developed that consists of a radon collector, a 60-l cylindrical buffer tank, three kinds of filter and an ionization chamber of flow-through type. The measured ionization current is continuously recorded in analogical form, then converted off-line into ^{222}Rn concentration using a calibration table derived in advance. From the ^{222}Rn concentration, the exhalation rate is obtained by calculation. The operating characteristics of this apparatus were determined from long-duration observations performed on the apparatus at different sites.

The apparatus is capable of providing an evaluation of the ^{222}Rn exhalation rate that is sufficiently accurate for all practical purposes.

2-2. Method of Measurement and Structure of Apparatus

The means adopted for measuring the exhalation rate of ^{222}Rn, is an improved version of Wilkening's method[1], and refers to others[2][3]. The ^{222}Rn collector is of structure and dimensions as shown in Fig. 2-1. At the collector entrance, an activated granular charcoal filter eliminates from the recycled air not only ^{222}Rn but also ^{220}Rn and their daughter nuclides that have been entrained in the preceding cycle.

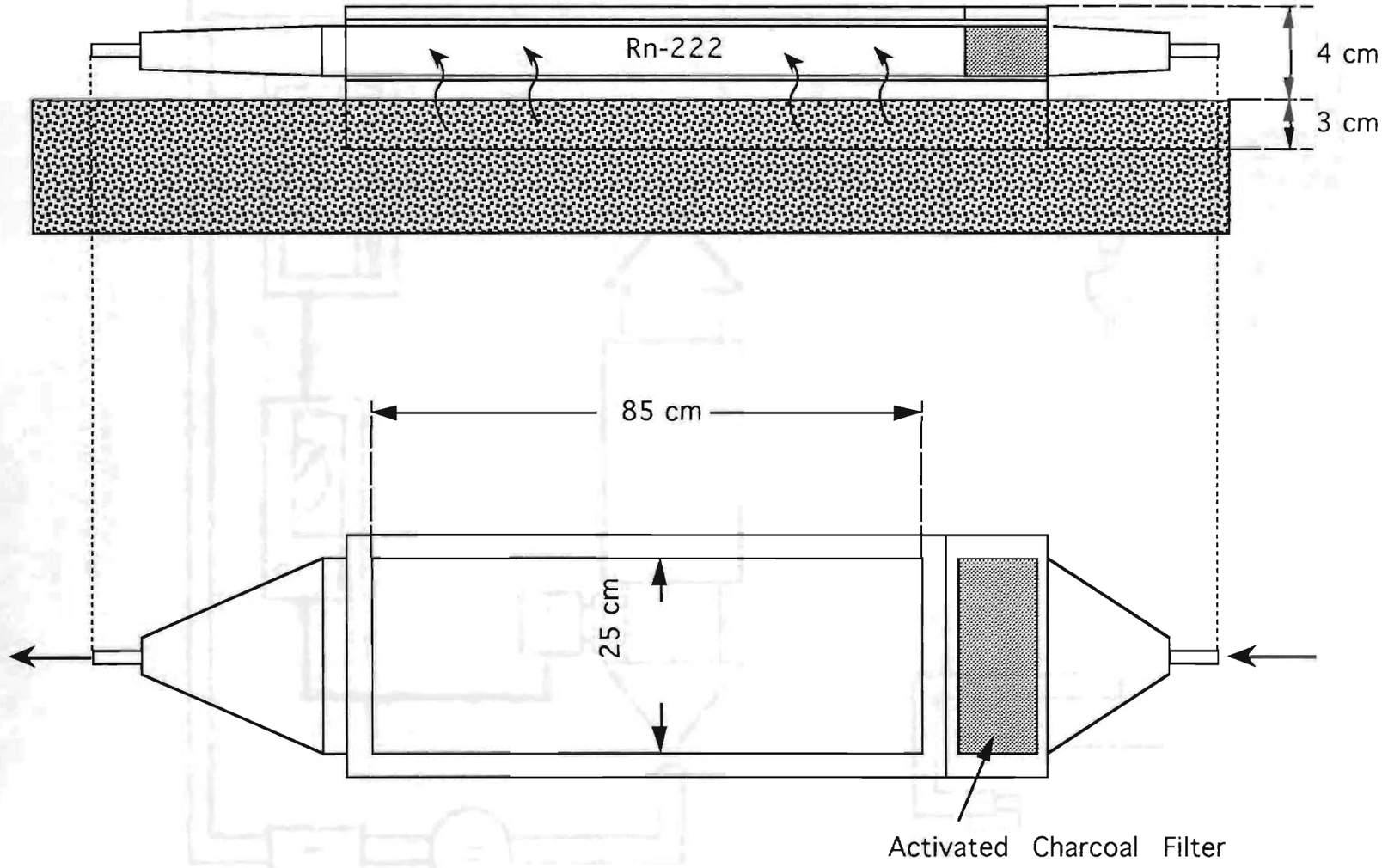


Fig. 2-1 Device for collecting ^{222}Rn exhalation

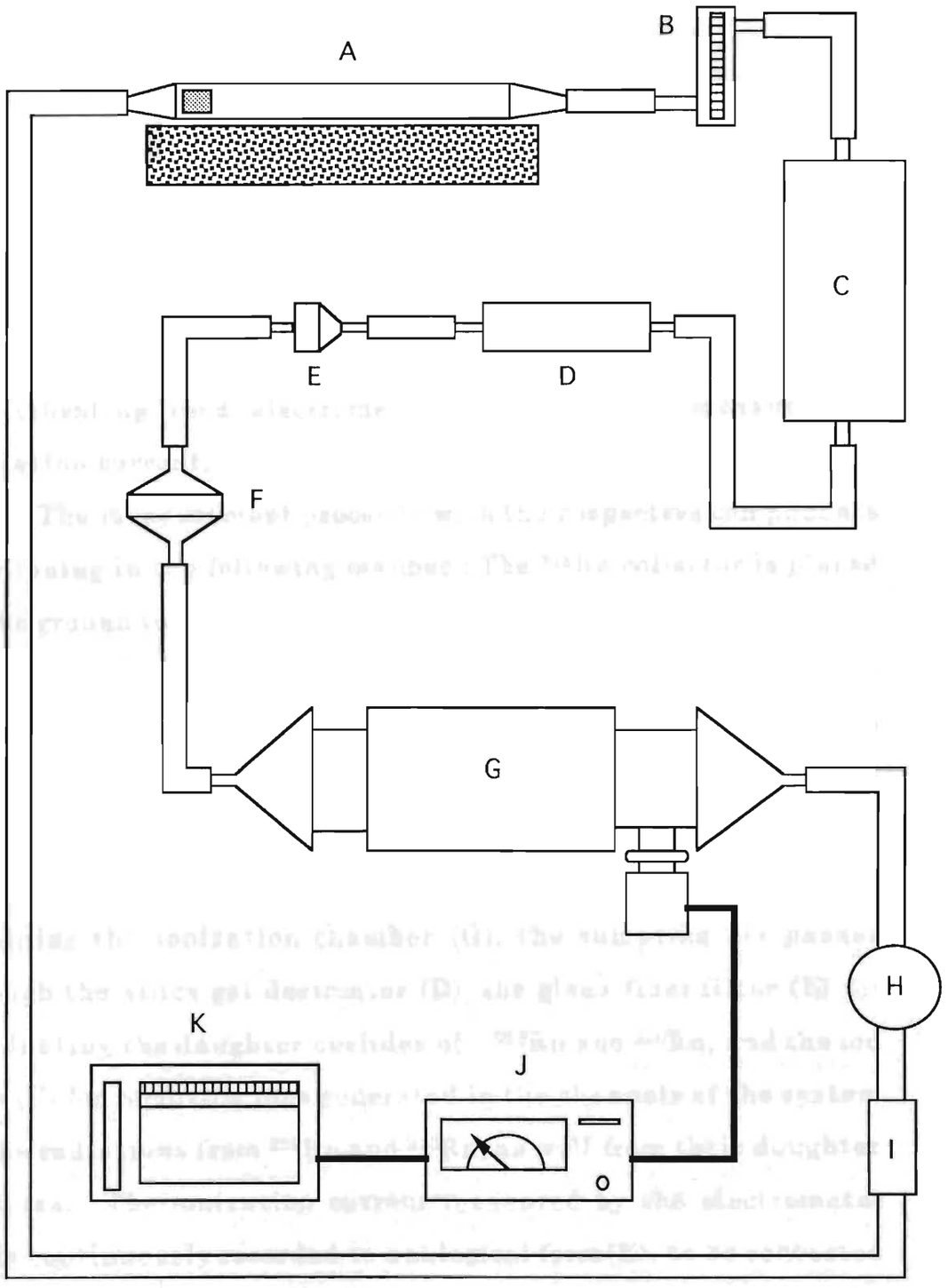


Fig. 2-2 Arrangement of apparatus for measuring ionization current generated by ^{222}Rn exhalation from ground.

The measuring systems is arranged as schematized in Fig. 2-2. The ionization chamber (G in Fig. 2-2) is cylindrical aspiration condenser with inner and outer cylinders measuring respectively 5 cm and 20 cm diameter and 45 cm in common length. The outer cylinder is maintained at a potential of -1,080 volts. The vibrating reed electrometer (J) is used to measure the ionization current.

The measurement proceeds with the respective components functioning in the following manner : The ^{222}Rn collector is placed on the ground to be measured ; the sampling air is adjusted to flow at 6 l/min, which is a rate that will not let the radon exhaled from ground be forcibly entrained ; this sampling air contained ^{222}Rn and ^{220}Rn is then passed into the 60-l buffer tank (C) to eliminate ^{220}Rn, whose half-life is only 54.5 sec and thus decays away during its stay in the buffer tank. Further downstream, before attaining the ionization chamber (G), the sampling air passes through the silica gel desiccator (D), the glass fiber filter (E) for eliminating the daughter nuclides of ^{222}Rn and ^{220}Rn, and the ion trap (F) for removing ions generated in the channels of the system by the radiations from ^{222}Rn and ^{220}Rn, as well from their daughter nuclides. The ionization current measured by the electrometer (J) is continuously recorded in analogical form(K), to be converted off-line into ^{222}Rn concentration by consulting a calibration table, and the resulting data are then used for calculating the ^{222}Rn exhalation rate, as described in the next section.

2-3. Deriving the ^{222}Rn Exhalation Rate from the Measured Ionization Current

For determining the relation between the measured ionization current and the concentration of entrained ^{222}Rn flowing through the ionization chamber, calibration measurements were made on radon gas exhaled from samples of sandy rock used for extracting titanium, containing in average 4,900 Bq/kg of ^{226}Ra.

Before proceeding on a calibration measurement, the initial zero point of ionization current was determined using Miranda's charcoal trap method[4]. Thereafter, the calibration proceeded as follows : Place a suitable quantity of the sandy rock sample in a spare 60-l tank ; connect this tank and the ^{222}Rn collector (A in Fig. 2-2) to the system ; close off the system and leave it standing until establishment of radioactive equilibrium in the tank between radon and its daughter nuclides, with ^{220}Rn decay away ; start up the pump (H in Fig. 2-2) to circulate the sampling air through the system, and let system as a whole ; adjust the pump speed to obtain a flow rate of 6 l/min ; while continuing to circulate the sampling at this rate through the system, measure the ^{222}Rn concentration in the tank, applying Thomas' two-filter method[5] ; plot the measured ^{222}Rn concentration against the corresponding reading of ionization current on the electrometer (J in Fig. 2-2) to obtain a calibration plot ; repeat this calibration measurement with the quantity of sandy rock parametrically varied, to generate the calibration curve.

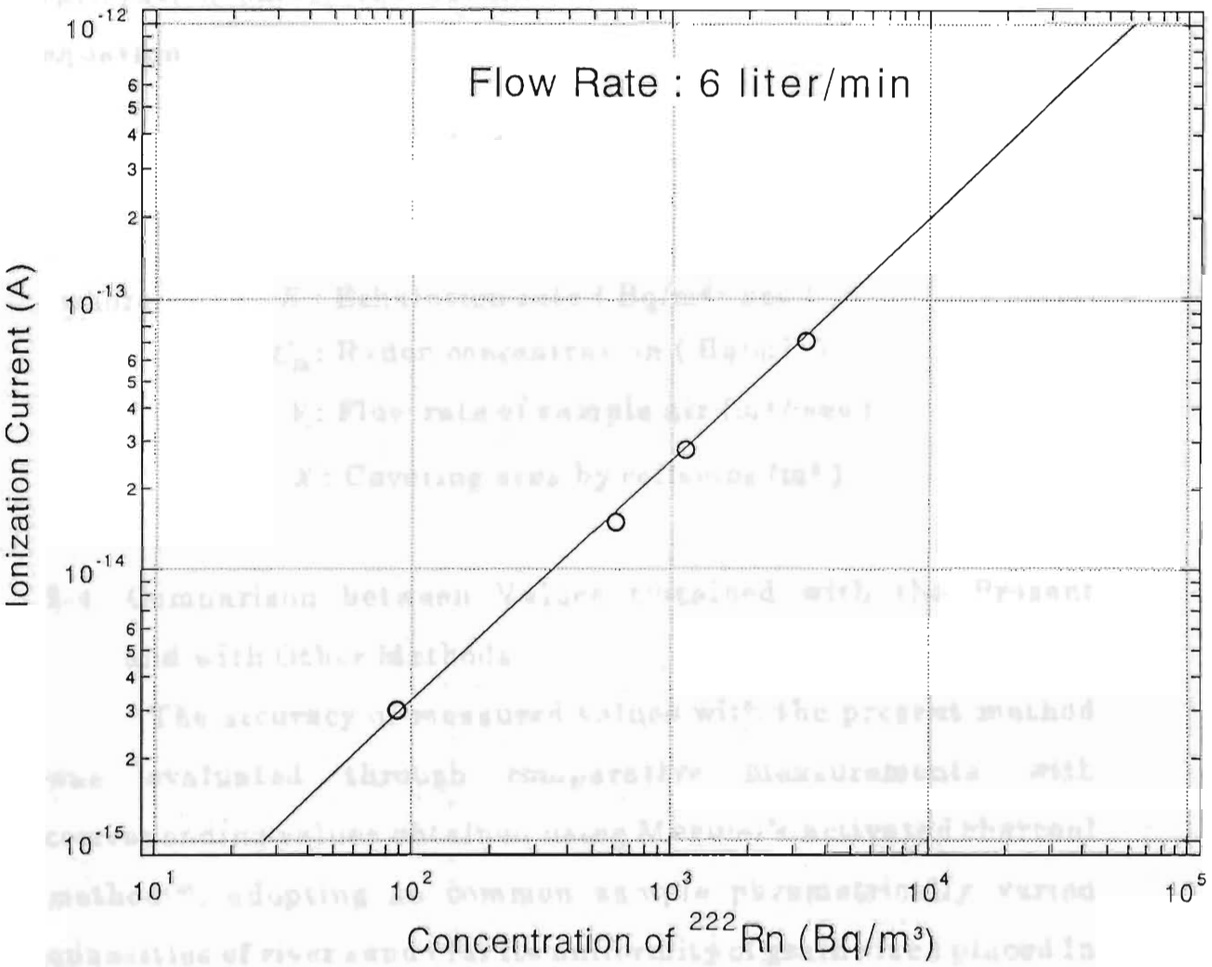


Fig. 2-3 Calibration plots relating measured ionization current to ^{222}Rn concentration

The calibration curve thus obtained is shown in Fig. 2-3, relating the ionization current to the ^{222}Rn in the sampling air flowing through the ionization chamber at 6 l/min. Using the values of ^{222}Rn concentration determined by means of this calibration curve, the exhalation rate is calculated with the equation

$$E = \frac{C_{Rn} \cdot V_s}{S}$$

where E : Exhalation rate ($Bq/m^2 \cdot sec$)

C_{Rn}: Radon concentration (Bq/m^3)

V_s: Flow rate of sample air (m^3 / sec)

S : Covering area by collector (m^2)

2-4. Comparison between Values Obtained with the Present and with Other Methods

The accuracy of measured values with the present method was evaluated through comparative measurements with corresponding values obtained using Megumi's activated charcoal method[8], adopting as common sample parametrically varied quantities of river sand (for its uniformity of grain size) placed in a wooden box measuring 2m × 2m × 2m. The measurements were performed inside a shed, to eliminate wind and other environmental effects. Experimental arrangement is shown in Fig. 2-4.

The results obtained from the comparative measurements are plotted in Fig. 2-5, from which a correlation coefficient of 0.91

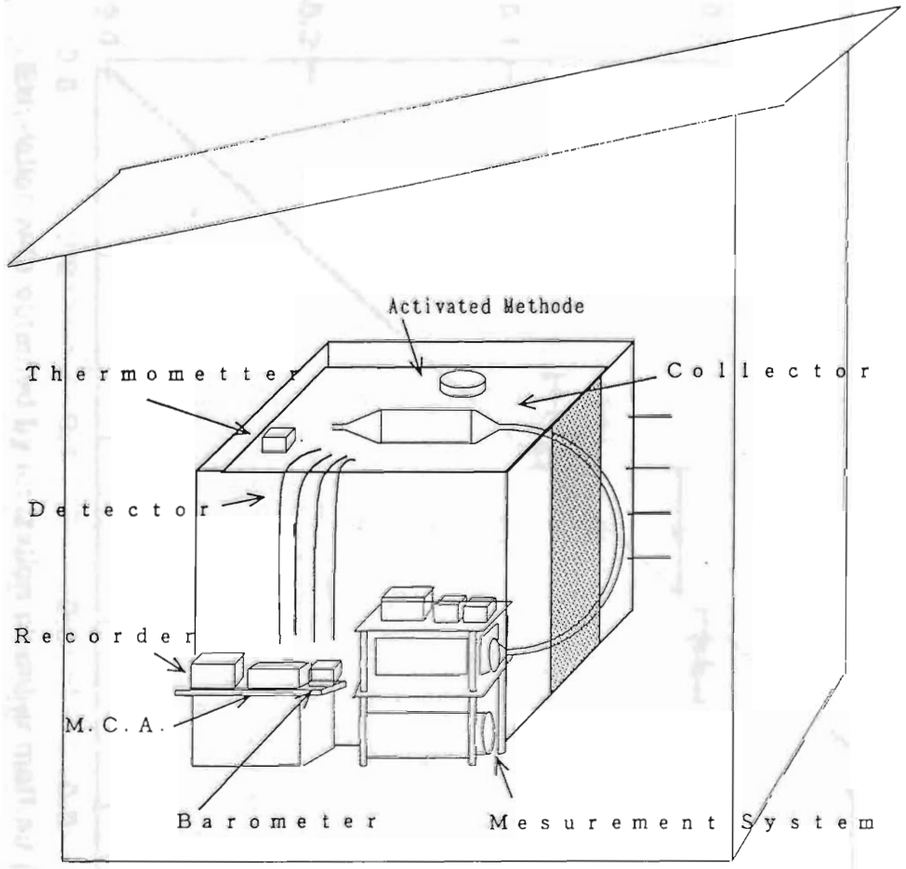


Fig. 2-4 Experimental arrangement for measuring ^{222}Rn exhalation and ^{222}Rn concentrations in air within soil composed of river sand.

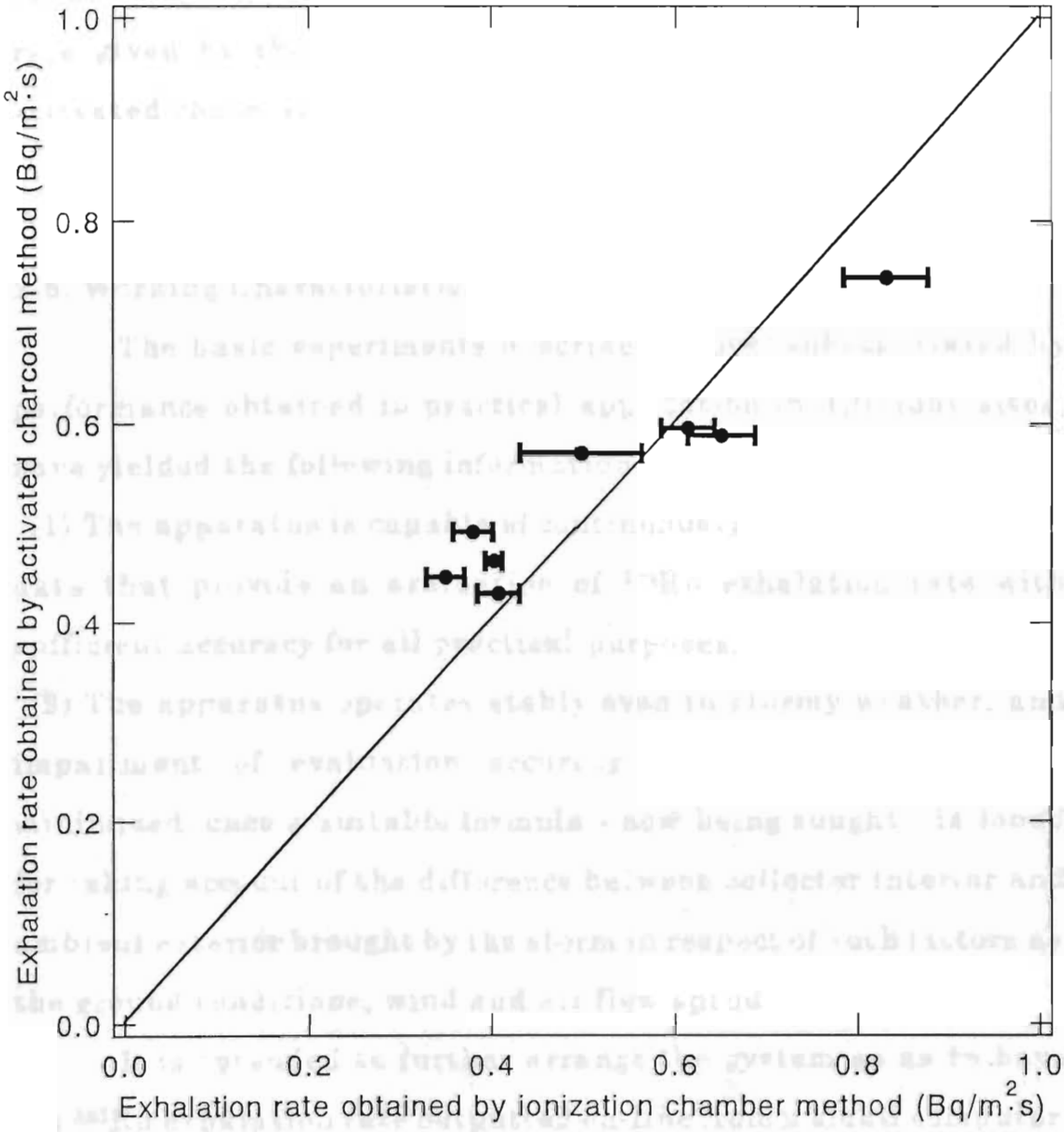


Fig. 2-5 Measured values of ^{222}Rn exhalation compared between present method and that using activated charcoal.

has been derived. The plots of Fig. 2-5 indicate a tendency toward slightly higher values in the higher range of exhalation rate given by the ionization chamber compared with those by activated charcoal method[7].

2-5. Working Characteristics

The basic experiments described above, substantiated by performance obtained in practical application in different sites, have yielded the following information :

(1) The apparatus is capable of continuously recording measured data that provide an evaluation of ^{222}Rn exhalation rate with sufficient accuracy for all practical purposes.

(2) The apparatus operates stably even in stormy weather, and impairment of evaluation accuracy can be expected to be minimized, once a suitable formula - now being sought - is found for taking account of the difference between collector interior and ambient exterior brought by the storm in respect of such factors as the ground conditions, wind and air flow speed.

It is intended to further arrange the system so as to have the ^{222}Rn exhalation rate outputted on-line from a small computer. Another matter that has not so far been considered is the effect, if any, brought on ^{222}Rn measurement by its adsorption onto the silica gel that is used for dehydrating the sampling air.

2-6. Practical Application of the Apparatus

a) Exhalation rate from the surface of an artificial ground

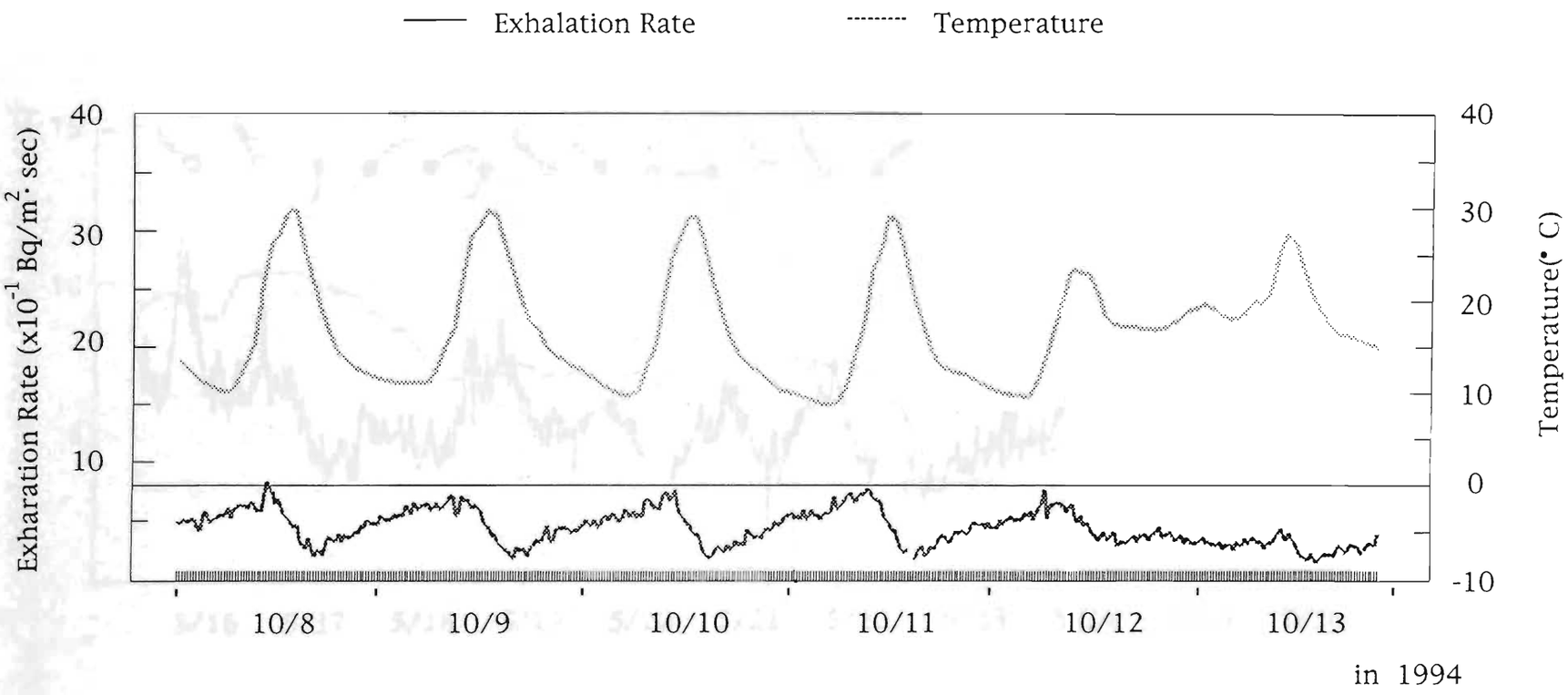


Fig. 2-6 An example of continuous recording of exhalation rate obtained on the artificial ground

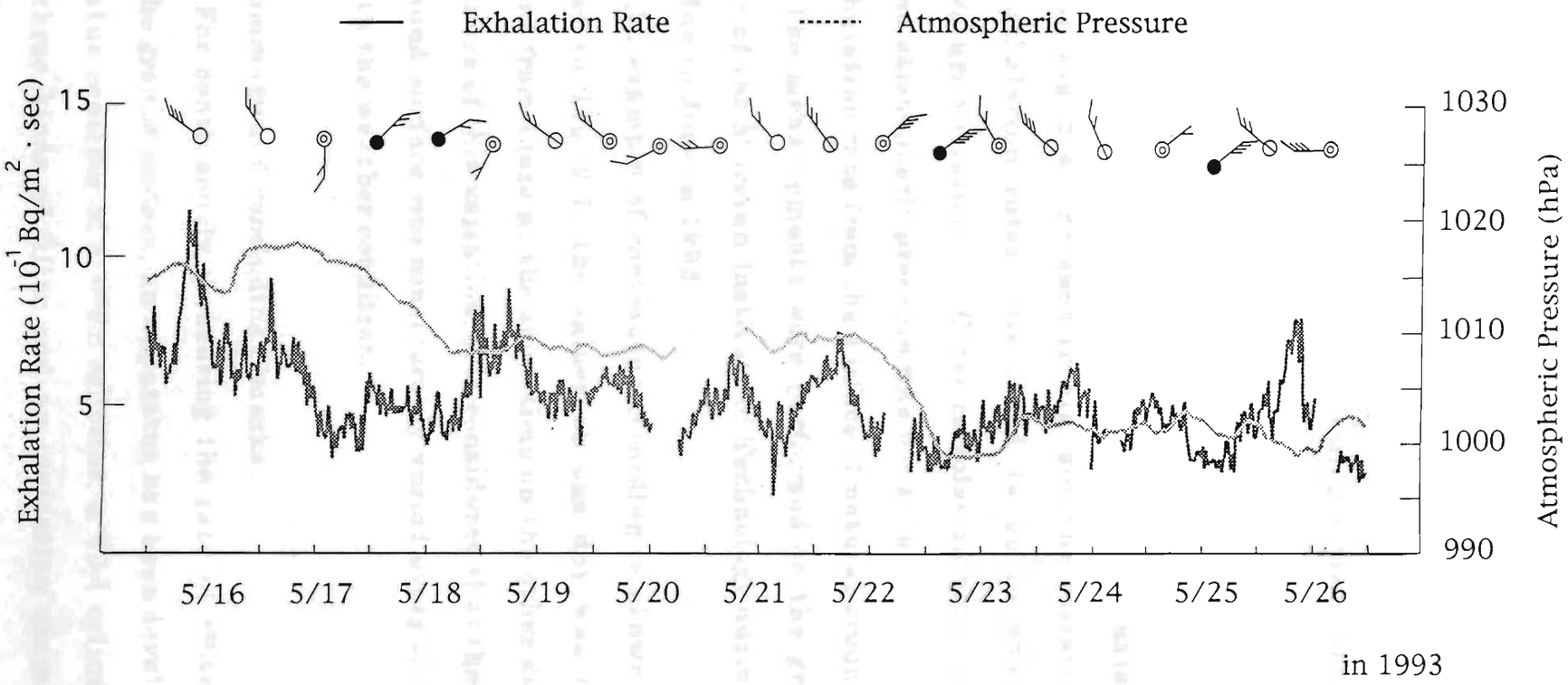


Fig. 2-7 An example of continuous recording of exhalation rate obtained on the natural ground

The river sand placed in the wooden box (described in paragraph 2-4.) was used for forming a hypothetical ground. The measurements were performed inside the shed to eliminate wind and rainfall during the period from June in 1994 to Feb. 1995.

An example of continuous recording of exhalation rate is shown in Fig. 2-6. As seen in Fig. 2-6, the variation trends of radon exhalation rates were found to be in phase with the temperature variations. Clear correlation between exhalation rate and atmospheric pressure was not seen.

b) Exhalation rate from the surface of natural ground

The measurements were performed on the ground at the campus of the Muroran Institute of Technology during the period from May to June in 1993.

An example of continuous recording is shown in Fig. 2-7. As seen in Fig. 2-7, the values on one day was considerably different from those at the same time on the other day. As the main cause of this variation, it is considered that the condition of the ground surface was considerably varied every time and every day with the weather condition. [8]

2-7. Summary and concluding remarks.

For continuously measuring the rate of ^{222}Rn exhalation from the ground surface, an apparatus has been developed. The apparatus consists of a ^{222}Rn collector, a 60-l cylindrical buffer tank, three kinds of filter and an ionization chamber of flow-through type.

To know the working characteristics and the accuracy of measured values, calibration measurements and comparison between values obtained with the presented and with other methods were made by applying two-filter method and by using activated charcoal method, respectively.

From the basic experiments described above and the practical use of the apparatus on the some different ground conditions, following informations were obtained ;

a) The apparatus is capable of continuously recording measured data that provide an evaluation of ^{222}Rn exhalation rate with sufficient accuracy.

b) The apparatus operates stably even in stormy weather.

c) From the measurements made on the ground inside of a house, the variation trends of exhalation rate were found to be in phase with the temperature variations.

d) From the measurements made on natural ground, the values on one day was considerably different from those at the same time on the other day. The ground condition affected with the weather seems to be main cause to effect variation of exhalation rates.

Chapter III

An apparatus for measuring ^{222}Rn progeny concentrations in atmosphere

III. An Apparatus for Measuring ^{222}Rn Progeny Concentrations in Atmosphere

3-1. Introduction

For the purpose of measuring the concentrations of ^{222}Rn progeny in the atmosphere, an apparatus has been developed that consists of a radon progeny collector, a silicon semiconductor detector and a pulse height analyzer. The operating characteristics of this apparatus were determined from long-duration observations performed at different sites.

The apparatus is capable of providing an evaluation of the ^{222}Rn concentrations and the ^{222}Rn progeny concentrations with sufficient accuracy.

3-2. Method of Measurement and Structure of Apparatus

The means adopted for measuring the concentrations of ^{222}Rn progeny, is an improved version of Mochizuki's method[1] and refers to others[2][3][4]. The ^{222}Rn progeny collector is of structure and dimensions as shown (a) and (b) in Fig. 3-1. A membrane filter (TOYO-ROSHI, Ltd. TM-300) was adopted for radon progeny collection. The silicon semiconductor detector (HORIBA, Ltd. Silicon Surface Barrier Type, 300SB 60L) was adopted for alpha ray detection of radon progenies collected on the filter. Characteristics of silicon semiconductor detector used are as follows ; surface area is 300 mm^2, effective useful thickness is 60 micro meter and resolution is 48.9 keV. The bias voltage used is 30 V. The detector output was fed to a multichannel

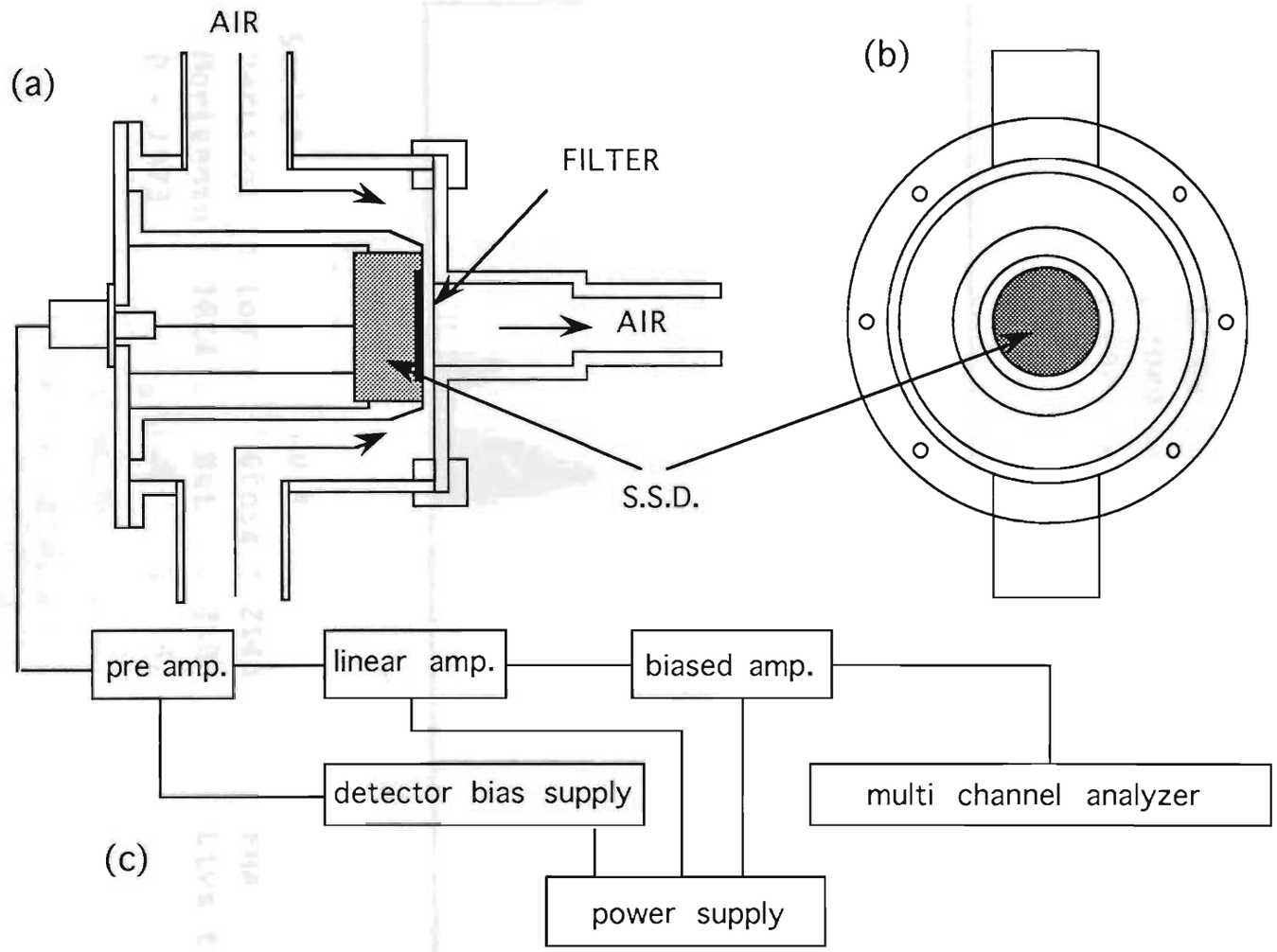
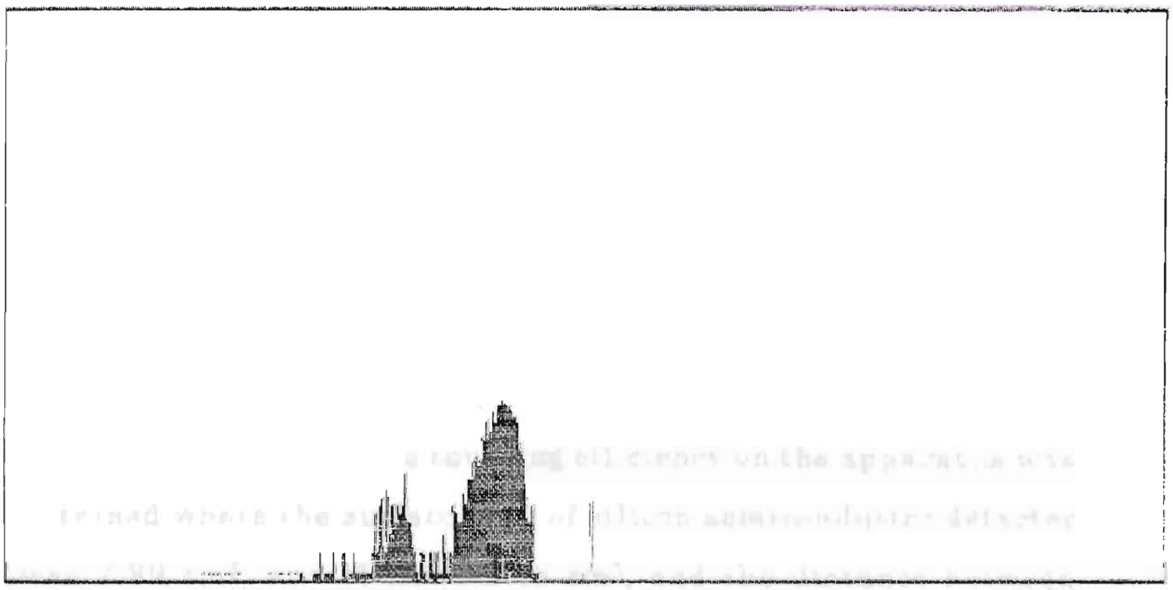


Fig. 3-1 Radioactive aerosol collector provided with silicon semiconductor detector and circuit on counting system for alpha ray spectrometry. (a)Horizontal cross section and (b)View of vertical cross section from the filter surface position (c)Electronic circuit on counting system.

15. Sep. '93 12:00
Live Time : 8000 sec
Real Time : 8000 sec



Scale*	ROI*	Pre Set*
Vertical : log	Gross : 2143	PHA
Horizontal: 1024	Net : 2100	Live time
0 - 1023	± : 47	

Fig. 3-2 An example of alpha ray spectrum obtained from ^{222}Rn progenies in the atmosphere.

analyzer that displayed relative integral alpha events as a function of energy. Alpha ray spectrometry was adopted for the measurement of ^{218}Po and ^{214}Po. Circuit used on counting system is as shown (c) in Fig. 3-1.

An example of alpha ray spectra obtained is shown in Fig. 3-2.

3-3. Counting Efficiency

The counting efficiency is calculated with equation(1-3) described in the chapter I paragraph 1-2-1.

$$[G] = \frac{1}{\pi d^2} \int_0^d \int_{-r}^{r} \frac{hx}{D(x)} \sqrt{\frac{E}{E + D(x)}} \, dg \cdot dx$$

A value 0.459 as a counting efficiency on the apparatus was obtained where the surface area of silicon semiconductor detector was 2.99 cm^2, and of filter 2.88 cm^2, and the distance between them was 0.3 cm, respectively.

3-4. Deriving the ^{222}Rn Concentration from the Measured Alpha-Counting Rate

Assuming radioactive equilibrium being established among ^{222}Rn, ^{218}Po, ^{214}Pb and ^{214}Bi, the time variation of alpha-counting on the filter with radioactive substances collected on it may be expressed as follows :

$$\lambda_A n_A = \lambda_B n_B = \lambda_C n_C$$

Regarding to ^{218}Po (alpha-ray counting of ^{218}Po)

$$I_{\alpha 1} = \lambda_A N_A = \varepsilon Q n_{A1}\left(1 - e^{-\lambda_A t}\right) = \varepsilon Q n_{A1}\Phi_1(t)$$

Regarding to ^{218}Po and ^{214}Bi (alpha-ray counting of ^{218}Po and ^{214}Po)

$$I_{\alpha 2} = \lambda_A N_A + \lambda_C N_C$$

$$= \varepsilon Q n_{A2}\left\{1 - e^{-\lambda_A t} + \left(1 + \frac{\lambda_A}{\lambda_B} + \frac{\lambda_A}{\lambda_C}\right)\cdot\left(1 - e^{-\lambda_C t}\right)\right.$$

$$+ \frac{\lambda_A{}^2\lambda_C}{\lambda_B(\lambda_A - \lambda_B)(\lambda_B - \lambda_C)}\cdot\left(e^{-\lambda_B t} - e^{-\lambda_C t}\right)$$

$$\left. - \frac{\lambda_B\lambda_C}{(\lambda_A - \lambda_B)(\lambda_A - \lambda_C)}\cdot\left(e^{-\lambda_A t} - e^{-\lambda_C t}\right)\right\}$$

$$= \varepsilon Q n_{A2}\Phi_2(t)$$

where,

$I_{\alpha 1}$: Alpha disintegration rate of ^{218}Po on the filter at time t (dps)

$I_{\alpha 2}$: Alpha disintegration rate of the summing up of ^{218}Po and ^{214}Po on the filter at time t (dps)

N_A , N_C : Number of ^{218}Po and ^{214}Bi atoms collected on the filter, respectively

λ_A , λ_B , λ_C : Decay constant of ^{218}Po, ^{214}Pb and ^{214}Bi, respectively (sec^{-1})

ε : Collection efficiency of filter to radioactive aerosols

Q : Flow-rate (cm^3/sec)

n_{A1} : Concentration of ^{218}Po atoms in air ($atoms/cm^3$) (obtained from alpha-ray spectrum measurement of ^{218}Po as shown in Fig. 3-2)

n_{A2} : Concentration of ^{218}Po atoms in air ($atoms/cm^3$) (obtained from alpha-ray total counting of ^{218}Po and ^{214}Po)

Then the concentration of ^{218}Po atoms in the air (C_{RaA1} and C_{RaA2}) expressed in Ci unit is estimated from the alpha disintegration rate $I_{\alpha 1}$ and $I_{\alpha 2}$ (experimental values) by using the equation ;

$$n_{A1} = \frac{1}{\varepsilon Q}\frac{I_{\alpha 1}}{\Phi_1(t)} \qquad \text{and} \qquad C_{RaA1} = \frac{\lambda_A n_{A1}}{3.7\times 10^{10}}$$

(from alpha-ray spectrum measurement of ^{218}Po)

$$n_{A2} = \frac{1}{\varepsilon Q}\frac{I_{\alpha 2}}{\Phi_2(t)} \qquad \text{and} \qquad C_{RaA2} = \frac{\lambda_A n_{A2}}{3.7\times 10^{10}}$$

(obtained from alpha-ray total counting of ^{218}Po and ^{214}Po)

If radioactive equilibrium state is being established like $\lambda_{Rn} n_{Rn} = \lambda_A n_{A1}$ and $\lambda_{Rn} n_{Rn} = \lambda_A n_{A2}$, two concentrations for n_{Rn} values, $(\lambda_A/\lambda_{Rn})n_{A1}$ and $(\lambda_A/\lambda_{Rn})n_{A2}$ were obtained. So that the ratio n_{A2}/n_{A1} should be equal to 1. Thus, the observed ratio is useful to determine the fraction of secular equilibrium.

The alpha decay events from the collecting activity were

detected throughout the sampling period. After the correction of overall counting efficiency, the ratio, R, for secular equilibrium conditions was calculated from time integrals of above collection functions, which express the relative growth of ^{218}Po and ^{214}Po on the filter as a function of sampling time.

A relative error, $\Delta R/R$, calculated from $R=n_{A2}/n_{A1}$ is given by the equation,

$$\left|\frac{\Delta R}{R}\right| = \left|\frac{En_{A1}}{n_{A1}}\right| + \left|\frac{En_{A2}}{n_{A2}}\right| ,$$

where, En_{A1} and En_{A2} are the errors of n_{A1} and n_{A2}, respectively.

As described above,

$$n_{A1} = \frac{1}{\varepsilon Q}\cdot\frac{I_{\alpha 1}}{\Phi_1(t)} , \qquad n_{A2} = \frac{1}{\varepsilon Q}\cdot\frac{I_{\alpha 2}}{\Phi_2(t)},$$

where, ε, Q, $I_{\alpha 1}$ and $I_{\alpha 2}$ are the measuring values, and $\Phi_1(t)$ and $\Phi_2(t)$ are the constant values. Therefore, the probable errors, En_{A1}, of n_{A1} and En_{A2}, of n_{A2} are given as follows ;

$$\frac{\partial n_{A1}}{\partial \varepsilon} = -\frac{1}{\varepsilon^2 Q}\cdot\frac{I_{\alpha 1}}{\Phi_1(t)}$$

$$\frac{\partial n_{A1}}{\partial Q} = -\frac{1}{\varepsilon Q^2}\cdot\frac{I_{\alpha 1}}{\Phi_1(t)}$$

$$\frac{\partial n_{A1}}{\partial I_{\alpha 1}} = \frac{1}{\varepsilon Q}\cdot\frac{1}{\Phi_1(t)}$$

accordingly, En_{A1} is,

$$En_{A1}{}^2 = \left(-\frac{1}{\varepsilon^2 Q}\cdot\frac{I_{\alpha 1}}{\Phi_1(t)}\right)^2 \varepsilon_\varepsilon{}^2$$

$$+\left(-\frac{1}{\varepsilon Q^2}\cdot\frac{I_{\alpha 1}}{\Phi_1(t)}\right)^2 \varepsilon_Q{}^2$$

$$+\left(\frac{1}{\varepsilon Q}\cdot\frac{1}{\Phi_1(t)}\right)^2 \varepsilon_{I_{\alpha 1}}{}^2$$

or

$$\left(\frac{En_{A1}}{n_{A1}}\right)^2 = \left(\frac{\varepsilon_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\varepsilon_Q}{Q}\right)^2 + \left(\frac{\varepsilon_{I_{\alpha 1}}}{I_{\alpha 1}}\right)^2 \qquad (3\text{-}1)$$

and for En_{A2} ,

$$\left(\frac{En_{A2}}{n_{A2}}\right)^2 = \left(\frac{\varepsilon_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\varepsilon_Q}{Q}\right)^2 + \left(\frac{\varepsilon_{I_{\alpha 2}}}{I_{\alpha 2}}\right)^2 \qquad (3\text{-}2)$$

where, ε_ε , ε_Q , $\varepsilon_{I_{\alpha 1}}$ and $\varepsilon_{I_{\alpha 2}}$ are the probable errors of ε , Q , $I_{\alpha 1}$ and $I_{\alpha 2}$, respectively.

The maximum deviation, from the mean, of the experimentally determined values for these errors of the instruments amounted to ± 2 percent, ± 3 percent, ± 8.6 percent, and ± 10.5 percent for ε , Q , $I_{\alpha 1}$ and $I_{\alpha 2}$, respectively. Now, substituting these values to the equation (3-1) and (3-2), $En_{A1}/n_{A1} = \pm 0.093$ and $En_{A2}/n_{A2} = \pm 0.111$ are obtained. Therefore, the

results obtained for $\Delta R/R$ were certainly correct to within 20.4 percent.

3-5. Practical Application of the Apparatus

- Application to the measurements of radon concentration in the atmosphere over the ocean -

In the atmosphere over the Pacific Ocean, some measurements of radon and its daughters have been made by ship since 1975 (Mochizuki, 1978, 1982 ; Mochizuki et al., 1981, 1982, 1984, 1985 ; Tanji et al., 1992, 1993)[5][6][7][8][9][10][11].

From July to September in 1993, the observations were performed over the Indian Ocean via the South China Sea and the Philippine Sea, on board the research vessel "Hakuho Maru", Ocean Research Institute, University of Tokyo[12].

The measurement was made at an interval of about 4 hours or 8 hours through the full period of the cruise. Collection time of radon daughters and analyzing time were both set at 8000 sec. Radon daughters were collected on a membrane filter (TOYO-ROSHI, Ltd. TM-100) with a suction pump at flow rate of 60 l/min. Sample air was introduced through a sampling hole of laboratory at height about 10 m above the sea surface.

Cruise routes of the expedition are shown in Fig. 3-3. Results obtained in each part on the routes are presented in Fig. 3-4 and Fig. 3-5.

Wind direction, wind force and other data obtained were referred by the meteorological instruments on the ship. Wind direction and wind force (in Beaufort wind scale) are shown in the

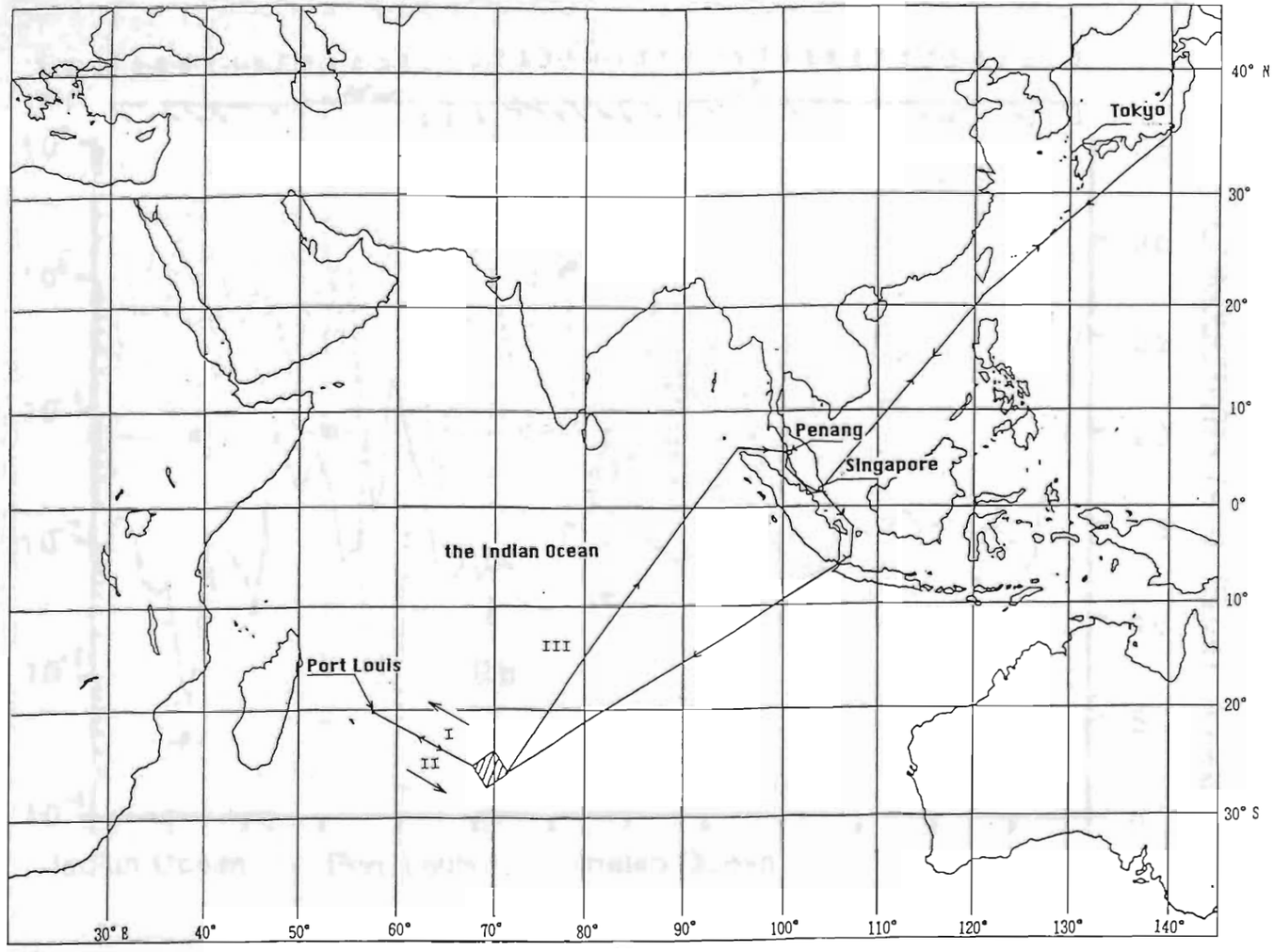


Fig. 3-3 Cruise route of KH 93-3 expedition, 8 Jul. to 17 Sep. 1993, by the research vessel Hakuho Maru, Ocean Research Institute, University of Tokyo.

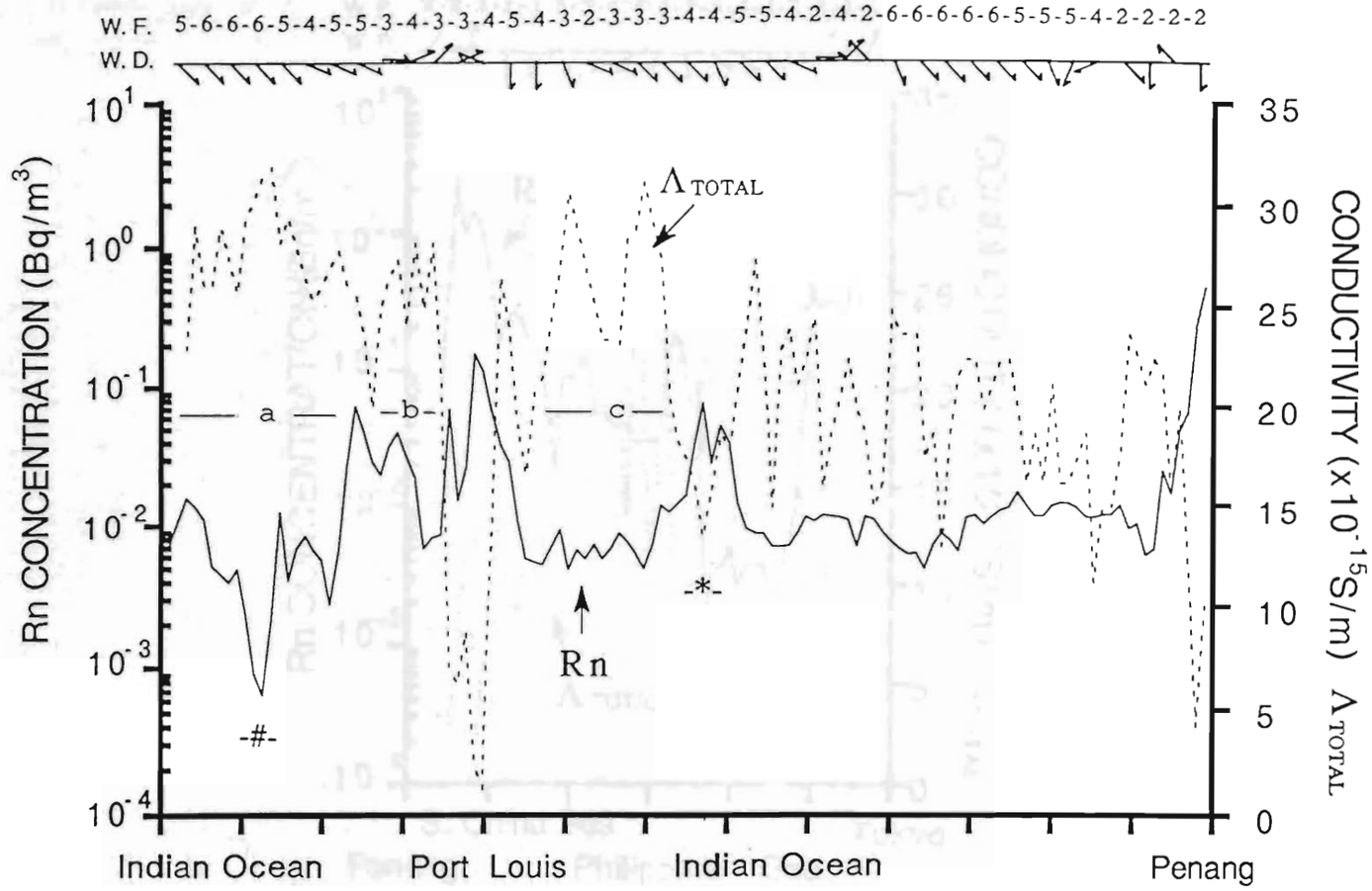


Fig. 3-4 ^{222}Rn concentration and electrical conductivity in the atmosphere over the Indian Ocean.

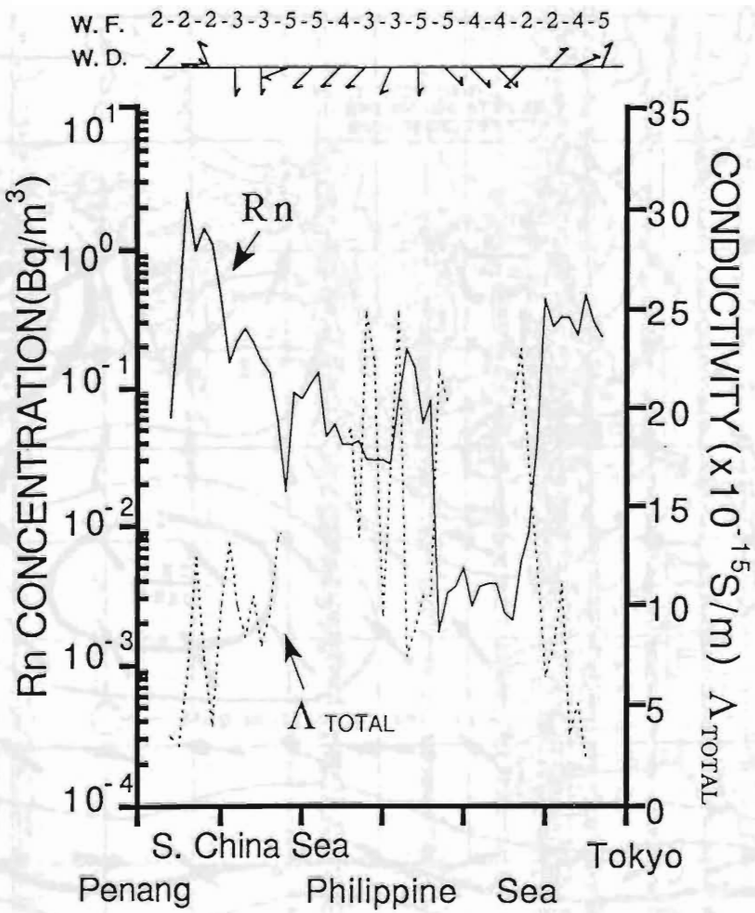


Fig. 3-5 ^{222}Rn concentration and electrical conductivity in the atmosphere over the South Chine Sea and Philippine Sea.

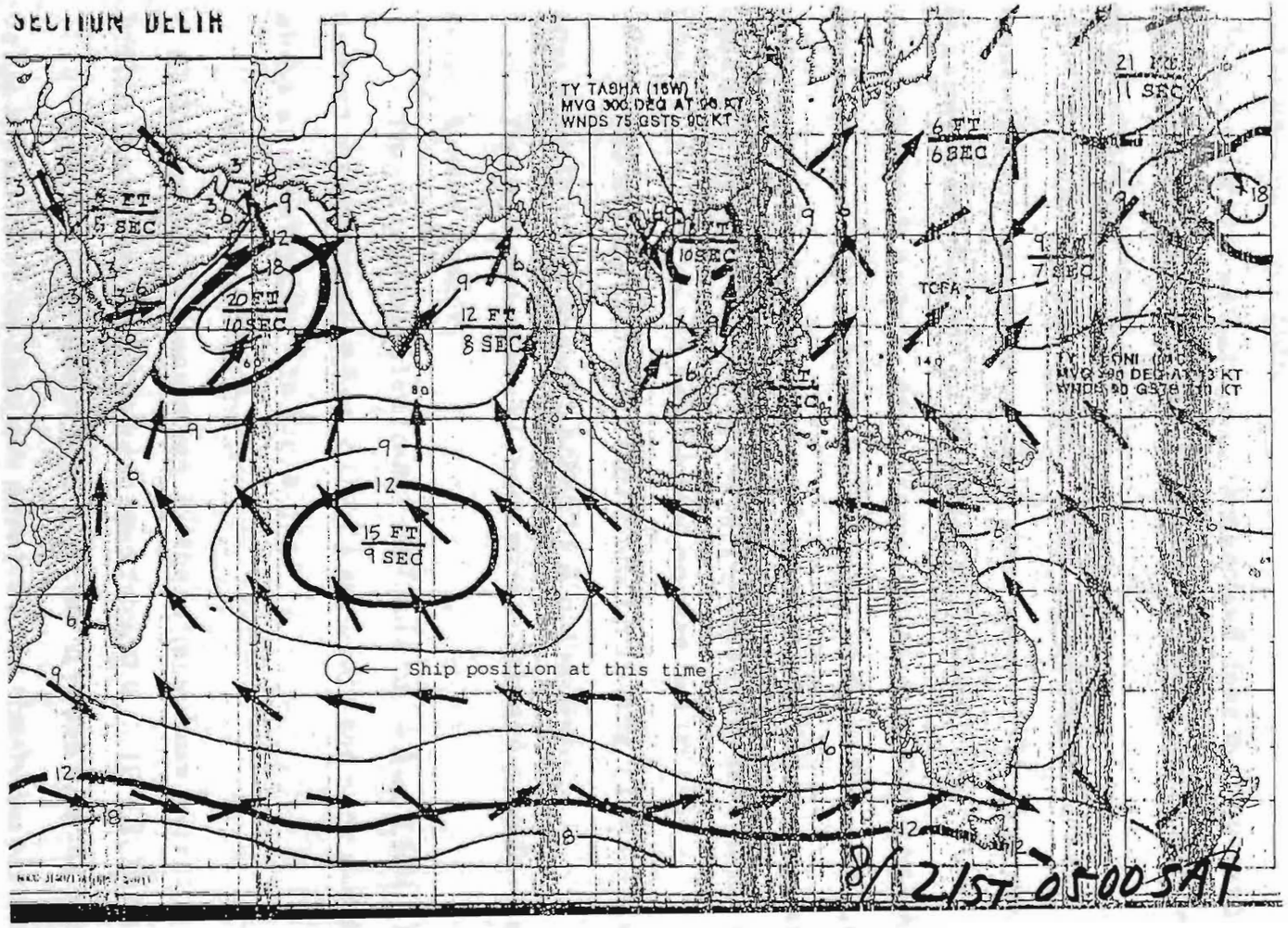


Fig. 3-6 Weather map of the Indian Ocean on 21 Aug. 1993.

upper part of each figure.

In Fig. 3-4 and Fig. 3-5 the abscissa does not represent the ship position on the cruise route but the time scale divided in equal interval of 2.5 days.

The values plotted were obtained at the time when each measurement was made on the cruise route. The ship position is known from the time on the abscissa.

Figure 3-4 shows the observation results obtained on the cruise routes I, II and III shown in Fig. 3-3. The continuous measurements of concentration of radon daughters and electrical conductivity were started on 1 Aug. (GMT) from around the spot (25° S, 69° E) shown as a shaded square in Fig. 3-3.

The vessel arrived at Port Louis on 10 Aug. (GMT) via the spot described above and marked by # in Fig. 3-4.

The measurements of them were continued also during the vessel was at anchor.

The vessel departed from Port Louis on 14 Aug. (GMT) and arrived at Penang on 2 Sep. (GMT) via again the spot described above and marked by * in Fig. 3-4.

As is seen in Fig. 3-4 ;

a) The radon concentration in the atmosphere over the mid Indian Ocean was found to be ranged from 6.6×10^{-4} Bq/m^3 to 7.6×10^{-2} Bq/m^3. The lowest concentration of radon and the highest value of electrical conductivity were found on the Ocean around the spot marked by # on 4 Aug. (GMT). The mean level of radon concentration measured during the cruise was about 1×10^{-2} Bq/m^3. This concentration level is similar to that obtained over

the south-eastern Pacific Ocean (Mochizuki et al., 1985 ; Tanji et al., 1992)[9][10].

b) Regardless of at the same spot on the Ocean shown in Fig. 3-3, variation patterns were different for different times as marked by # and * in Fig. 3-4. In the observation from 3 Aug. to 4 Aug. (mark #), abrupt decrease of radon was seen, although its concentration level was extremely low, and electrical conductivity showed considerably high values. While, in the observation from 19 Aug. to 21 Aug. (mark *), considerable increase and decrease of radon, although its concentration level was still fairly low, were seen and the electrical conductivity showed the abrupt change between low and high values corresponding to radon increase and decrease.

Referring to the weather map shown in Fig. 3-6 and considering the wind directions shown in Fig. 3-4, the increase of radon measured from 19 Aug. to 21 Aug. seemed to be under the considerable influence from Australian Continent.

At the present, detailed analysis of air mass trajectory has not yet been made. However, roughly speaking, the extremely low level of radon concentration measured from 3 Aug. to 4 Aug. is thought to be due to the aged air mass that had passed long over the Antarctic Ocean.

Clear inverse correlation between radon concentration and electrical conductivity was seen in the areas (a, b and c in Fig. 3-4) over the Ocean from vicinity of the spot (# in Fig. 3-4) to Port Louis and from Port Louis to the spot (*) .

Figure 3-5 shows the observation results obtained on the

cruise route from Penang to Tokyo.

As it is seen on the cruise route shown in Fig. 3-3, many islands are scattered along the route and also Asian Continent lies on the left side of the vessel. Corresponding to this, the concentration of radon and electrical conductivity showed considerable variations when the vessel approached to and departed from the vicinity of islands.

The concentration of radon measured here clearly showed a considerable influence from islands and Asian Continent. The radon concentration was ranged from 1.7×10^{-2} Bq/m^3 to 1.9×10^{-1} Bq/m^3, and the mean concentration level of radon was higher than that obtained over the Indian Ocean.

In the observation period from 11 Sep. to 13 Sep. when the vessel was on the Philippine Sea, for escape from the typhoon just taking place over the sea of south of Formosa, the vessel cruised along the east side of typhoon at a considerable distance departed from the scheduled route.

The radon concentration measured on this occasion is thought to be from the air mass that had passed over the Pacific Ocean, and it showed a considerably low level from 1.7×10^{-3} Bq/m^3 to 5.0×10^{-3} Bq/m^3.

Observational evidences obtained from the measurements performed over the ocean are summarized as follows ;

a) on the mid Indian Ocean, radon concentration levels were found to be ranged from 6.6×10^{-4} Bq/m^3 to 7.6×10^{-2} Bq/m^3.

b) The lowest value of radon concentration, 6.6×10^{-4} Bq/m^3, was found around the spot 25° S, 69° E.

c) On the South China Sea, the radon concentration level was ranged from 1.7 $\times 10^{-2}$ Bq/m^3 to 1.9 $\times 10^{-1}$ Bq/m^3.

d) Clear inverse correlation between radon concentration and electrical conductivity was found in the observation over the mid Indian Ocean.

3-6. Summary and concluding remarks

For measuring the concentrations of ^{222}Rn progeny in the atmosphere, an apparatus has been developed. The apparatus consists of a ^{222}Rn progeny collector, a silicon semiconductor detector and a pulse height analyzer. A membrane filter (TOYO-ROSHI, Ltd. TM-300) was adopted for ^{222}Rn progeny collection. A silicon semiconductor detector (HORIBA, Ltd. 300SB 60L) was adopted for alpha ray detection of ^{222}Rn progenies collected on the filter. Alpha ray spectrometry was adopted for the measurements of ^{218}Po and ^{214}Po. As a counting efficiency of this apparatus, a value 0.459 was obtained. A relative error was estimated to be within 20.4 percent.

From the observations made in the atmosphere over the ocean, it was confirmed that the apparatus is useful to measure the extremely low level concentrations of ^{222}Rn progeny with sufficient accuracy.

Observational evidences obtained from the measurements performed over the Indian Ocean are as follows ;

a) On the mid Indian Ocean, radon concentration levels were found to be ranged from 6.6 $\times 10^{-4}$ Bq/m^3 to 7.6 $\times 10^{-2}$ Bq/m^3 and the values of electrical conductivity were from 1.3 $\times 10^{-14}$ S/m to 3.1

$\times 10^{-14}$ S/m.

b) The lowest value of radon concentration, 6.6 $\times 10^{-4}$ Bq/m^3 , was found around the spot 25° S, 69° E.

c) On the South China Sea, the radon concentration level was ranged from 1.7 $\times 10^{-2}$ Bq/m^3 to 1.9 $\times 10^{-1}$ Bq/m^3.

d) Clear inverse Correlation between radon concentration and electrical conductivity was found in the observation over the mid Indian Ocean.

Chapter IV

An apparatus for measuring unattached ^{218}Po in atmosphere

VI. An Apparatus for Measuring Unattached ^{218}Po Concentration in Atmosphere

4-1. Introduction

Speaking of the branch of health physics, the normal concentration of radon found in the atmosphere does not seem to bring up severe problems on health hazard. Despite this, it may be stressed from the basis of animal studies and human experiences that inhalation of radon and its daughters is probably the source of chief health hazard in the mining or the radon concentrated room air.(1)

For assessing the deposition characteristics of inhaled radon daughter products, it is firstly needed to know the relative amount of airborne daughters in the unattached state and secondarily the size distributions of attached state.

For the purpose of measuring the concentration of unattached ^{218}Po in the air, an apparatus has been developed that consists of a ^{218}Po collector with a wire mesh filter, a semiconductor detector, and a pulse height analyzer.

The apparatus is capable of providing an evaluation of the unattached ^{218}Po concentration and the ratio of unattached ^{218}Po to attached ^{218}Po, simultaneously.

4-2. Method of Measurement and Structure of Apparatus

The Unattached ^{218}Po has been measured with a diffusion tube or a wire screen. The wire screen method was applied by James et al., 1972(1), Thomas et al., 1972(2), and the others(3)(6).

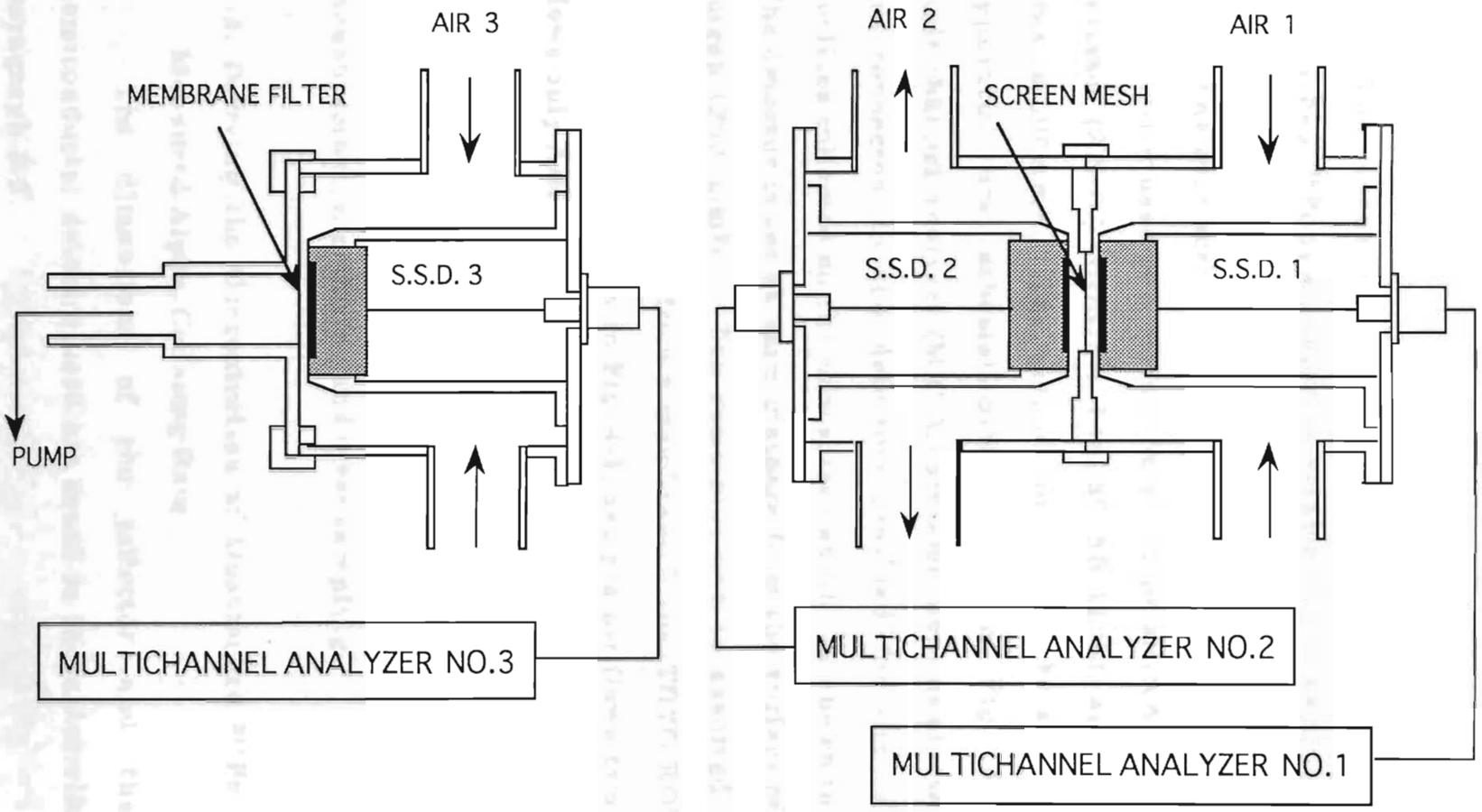


Fig. 4-1 Arrangement of apparatus for measuring unattached ^{218}Po concentration and attached ^{218}Po concentration.

The means adopted for measuring the concentration of unattached ^{218}Po is an improved version of, James's method[2] and Thomas's method[3].

The wire screen used is made of metal and has 300 mesh. The detector used is a surface barrier type silicon semiconductor detector (S.S.D.) (HORIBA, Ltd. 300SB-120L), and its effective area is 300mm^2. The method applied and the structure of the apparatus are schematically shown in Fig. 4-1. Three multichannel analyzer (M.C.A.) systems were used, two of them are connected to the detectors provided for the radioactive nuclides collected on the wire screen as like as shown in Fig. 4-1. The detector is set at 3mm distance from the surface of the wire screen (300 mm^2). The remaining one is assorted with the detector set at 3mm from a membrane filter (TOYO ROSHI, Ltd. TM-100). As shown in Fig. 4-1, sample air flows two way, one flows only AIR 3 and the other flows through AIR 1, 2 and 3.

The unattached fractions of ^{218}Po are determined from the simultaneous alpha ray spectrometric analysises with three M.C.A. Alpha ray spectrometric analysises are made twice a measurement, i. e. during and after sampling.

4-3. Deriving the Concentration of Unattached ^{218}Po from the Measured Alpha-Counting Rate

The dimensions of the collector and the silicon semiconductor detector used are equal to those described in the paragraph 3-2.

The collection efficiency of the wire mesh filter on the

unattached ^{218}Po and appearing efficiency of ^{218}Po collected on the filter were referred the data obtained by James[1] and adopted 0.72 and 0.686, respectively.

As the counting efficiency of the system, the same value, 0.459, described in the paragraph 3-3 was applied because the distance between the wire mesh screen and the semiconductor detector was set up equal to the condition in the case of the measurement of radon concentration.

The relative errors involved in measured values were evaluated with the same manner as those described in the paragraph 3-4.

Referring the consideration on the collection efficiency of the wire mesh filter to attached ^{218}Po by Shimo[4], the collection efficiency on attached ^{218}Po was neglected on account of its low value of 1% or less.

4-4. Practical Application of the Apparatus.

Some observation results of unattached ^{218}Po are shown in Fig. 4-2. They are obtained from the measurements carried out at the basement of the Oshamanbe campus of the Science University of Tokyo from July 23 to 25, in 1989. These measurements were made at 600 cm^3/sec for sample air flow rate and measuring time 1,000 sec, with growth method of alpha ray spectrometry.

In Fig. 4-2, an ordinate represents the percentage of unattached ^{218}Po concentration to all attached ^{218}Po. In Fig. 4-3, an ordinate represents the ratio of unattached ^{218}Po

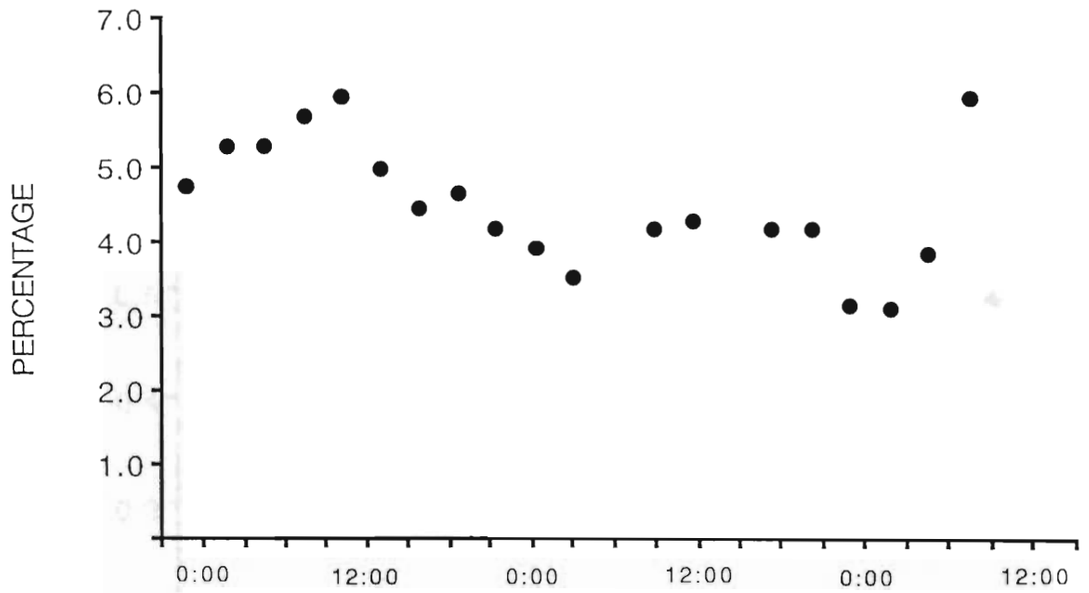


Fig. 4-2 Variation of the ratio of unattached ^{218}Po concentration to ^{218}Po concentration.

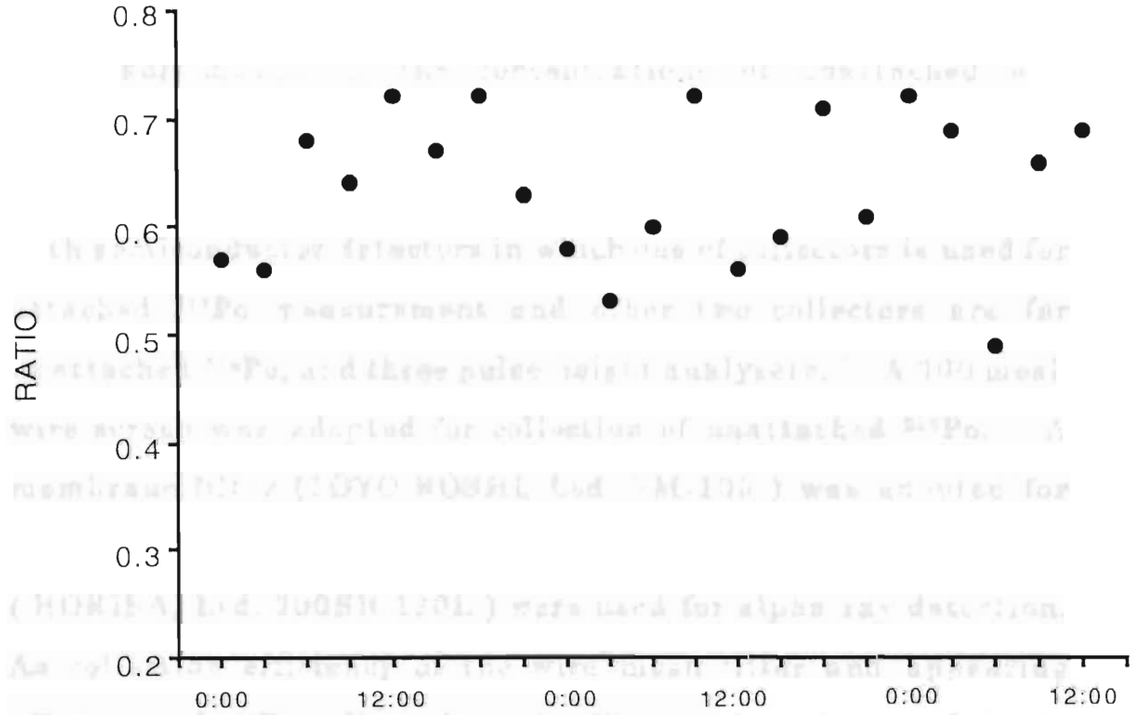


Fig. 4-3 Characteristics of the wire mesh filter on the collection of unattached ^{218}Po. The plots show the ratio of unattached ^{218}Po concentration obtained on the reverse side of the filter to that on the front side.

concentration on the reverse side of mesh to the unattached ^{218}Po on the front side of mesh. Each data show a considerable variation, but about these phenomena and their cause, detail discussion is a future subject.

4-5. Summary and concluding remarks

For measuring the concentrations of unattached and attached ^{218}Po simultaneously, an apparatus has been developed. The apparatus consists of three kinds of ^{218}Po collector provided with semiconductor detectors in which one of collectors is used for attached ^{218}Po measurement and other two collectors are for unattached ^{218}Po, and three pulse height analyzers. A 300 mesh wire screen was adopted for collection of unattached ^{218}Po. A membrane filter (TOYO-ROSHI, Ltd. TM-100) was adopted for collection of attached ^{218}Po. Silicon semiconductor detectors (HORIBA, Ltd. 300SB 120L) were used for alpha ray detection. As collection efficiency of the wire mesh filter and appearing efficiency of ^{218}Po collected on the filter, values 0.72 and 0.686 were adopted in present work, respectively.

From some measurements carried out in a basement air, it was confirmed that the apparatus is capable of providing an evaluation of the unattached ^{218}Po concentration and the ratio of unattached ^{218}Po to attached ^{218}Po, simultaneously.

ACKNOWLEDGMENT

ACKNOWLEDGMENT

I would like to express my sincere gratitude to Professor Sadamu Mochizuki, Muroran Institute of Technology, for his guidance in this work. I would also like to thank Assistant Professor Tatsuo Tanji, Muroran Institute of Technology, for his kind encouragement and cooperation.

The author also thanks to the graduate students, Tsuguaki Itaya, Yoichi Hiruta and Yutaka Sato, for their assistance to measurements.

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FIGURES and TABLES

FIGURES AND TABLES

TABLES

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Tab. 1-2 Diffusion constants in air of soil constructed from river sand.

FIGURES

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Chapter 3

Fig. 3-1 Radioactive aerosol collector provided with silicon semiconductor detector and circuit on counting system for alpha ray spectrometry. (a) Horizontal cross section and (b) view of vertical cross section from the filter surface position.

Fig. 3-2 An example of alpha ray spectrum obtained from ^{222}Rn progenies in the atmosphere.

Fig. 3-3 Cruise route of KH 93-3 expedition, 8 Jul. to 17 Sep. 1993, by the research vessel Hakuho Maru, Ocean Research Institute, University of Tokyo.

Fig. 3-4 ^{222}Rn concentration and electrical conductivity in the atmosphere over the Indian Ocean.

Fig. 3-5 ^{222}Rn concentration and electrical conductivity in the atmosphere over the South China Sea and the Philippine Sea

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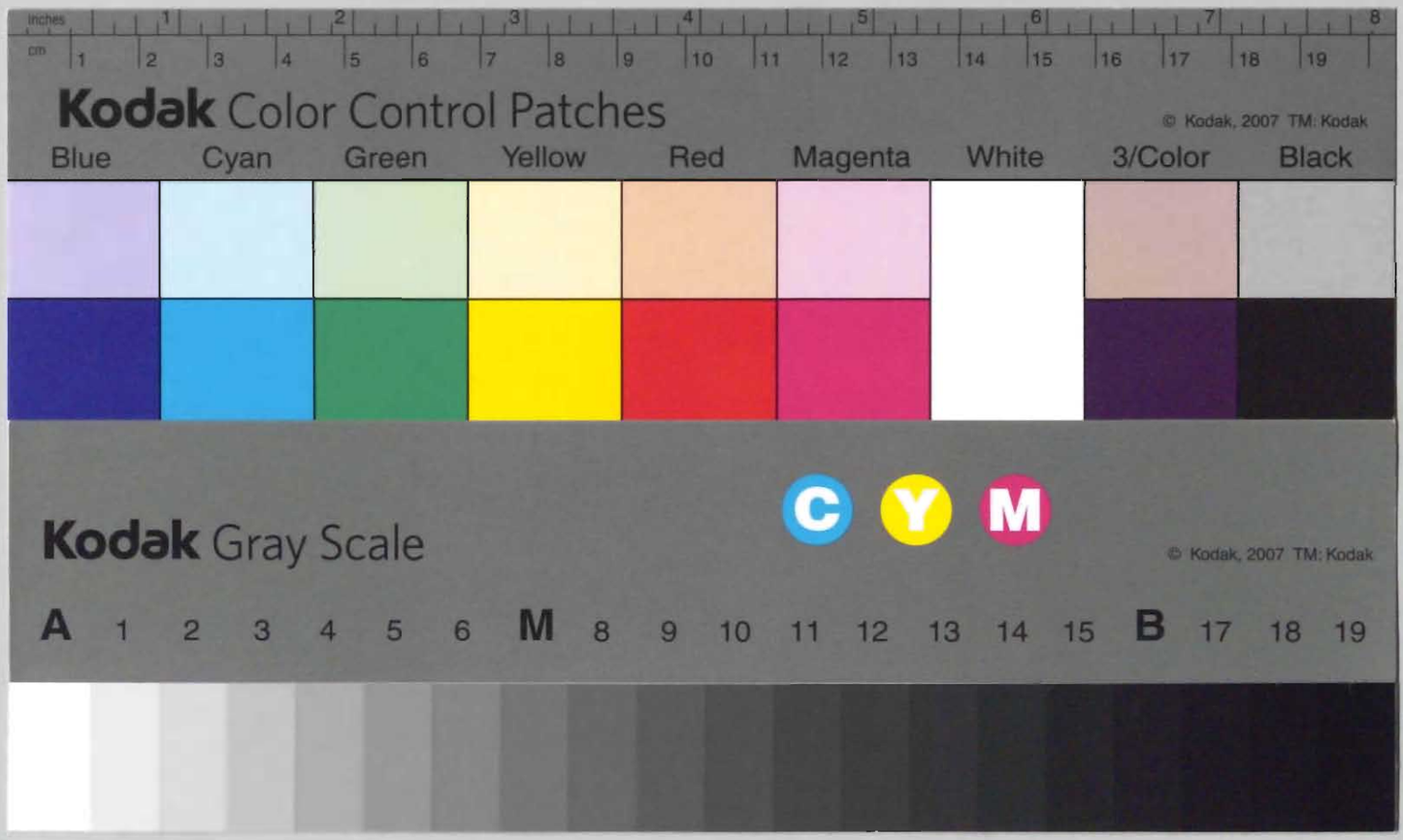
Fig. 4-2 Variation of the ratio of unattached ^{218}Po concentration to ^{218}Po concentration.

Fig. 4-3 Characteristics of the wire mesh filter on the collection of unattached ^{218}Po. The plots show the ratio of unattached ^{218}Po concentration obtained on the reverse side of the filter to that on the front side.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